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THE DEMONSTRATION OF THE FEASIBILITY OF THE TUNING AND STIMULATION OF NUCLEAR RADIATION

Short Title: GAMMA-RAY LASER

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This project concerns the demonstration of the feasibility of the tuning and perhaps even stimulation of nuclear radiation. Theory has indicated that anti-Stokes Raman upconversion of intense but conventional long wavelength sources of radiation produced by scattering from isomeric states of nuclear excitation could lead to significant sources of

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20. Abstract (continued)

tunable γ-radiation characterized by the natural Mössbauer widths of the lines. This would result in lines with sub-Angstrom wavelengths and widths of a few MHz. Whether or not these processes can reach threshold depends upon the resolution of basic issues lying in an interdisciplinary region between quantum electronics and nuclear physics that have not been previously addressed. It was the purpose of this work to study these issues experimentally.

The overall problem being addressed is a broad one that naturally divides into three lines: 1) Coupling pump power into a nucleus, 2) Extracting nuclear excitation with a radiation field, and 3) Insuring material survival. Such a perspective is beyond the scope of a single project and only the second line has been addressed by the work supported by this grant.

During this three-year period major progress was realized in the extraction of nuclear excitation with a radiation field. Breakthrough work continued to show that spin waves propagating in a host medium could affect nuclear properties that determine how well nuclear excitation can be coupled to the radiation field. These feasibility studies were completed with an analysis of the means for exploiting such effects to develop amplification without absorption at the nuclear level.

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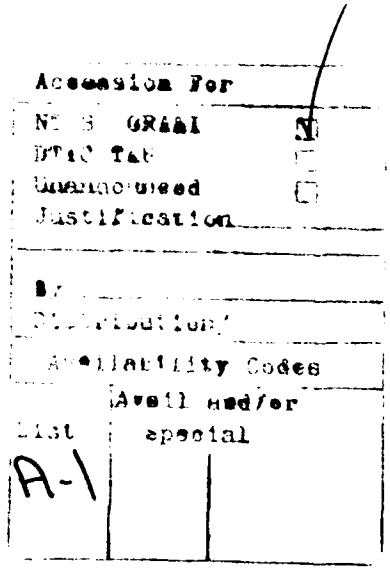

INTRODUCTION

The overall objective of our work is to demonstrate the feasibility of a gamma-ray "laser." At such wavelengths even the weakest coherent source would be of revolutionary importance, as it would enable the exploitation of the wave properties of gamma radiation. Pursuit of that goal was begun with ONR sponsorship of a contract preceding this grant. That first phase of that research into the question of the feasibility of a gamma-ray laser was concerned with problem definition. Evaluations of a number of options finally identified the nuclear analog of the ruby laser as the most likely vehicle for the realization of a gamma-ray laser.[1] The problems were identified as being threefold:

1) Coupling pump power into the nucleus,

2) Extracting nuclear excitation with the radiation field, and

3) Insuring material survival.

Because of the difficulties in developing support for such a breadth of effort in a single activity, the problem was divided roughly along these lines.

The challenge of coupling an intense pulse of some type of pump radiation into the nuclei of a material was spun-off to SDIO. Benefiting from access to large nuclear simulators such as DNA/Aurora at the Harry Diamond Laboratories and to the most intense linacs such as the superconducting injector to the storage ring at Darmstadt the pump mechanism was demonstrated.

A first breakthrough[2] showed that the energy stored in a long-lived nuclear isomer, $^{180}Ta^{m}$ could be dumped into fluorescence with an intense pulse of x rays by "optically pumping" it through a giant resonance with an integrated cross section approaching 10^{-21} cm^2 eV. Such a value is about 1000 times more favorably large than what had been measured for any previous nuclear fluorescence. It represented the discovery of a new class of excitation resonances

associated with some collective motion of the nucleus, as yet uncharacterized by theory. In a second breakthrough[3] it was shown experimentally that this giant pumping resonance lay at a favorably low excitation energy of 2.8 MeV. *We now know how to pump large amounts of excitation into nuclear systems* and the thrust along the first line of investigation is turning to the problems of nuclear systematics. The most recent experiments have used four accelerators to survey 19 isotopes for the systematic occurrence of these giant pumping resonances. The best material to pump must be identified.

Now that the nuclei have been pumped (or dumped) the second line of problems becomes of paramount importance. Instead of ending as nuclear fluorescence or as internal conversion, the nuclear excitation must be coupled to the radiation field for controlled extraction. For quite some time we have proposed[4] the use of resonant, longer wavelength radiation to mix the properties of the nuclear state being pumped by the primary process with properties of another state radiating more freely. In this way the lifetime of the state accumulating the population can be suddenly switched to a shorter value. Very recently Steve Harris has proposed the brilliant concept[5] of gain without inversion which can be readily adapted to the same type of nuclear environment. Critical to either approach is the ability to affect nuclear populations and to prepare initial quantum states while maintaining natural lifetimes under Mössbauer conditions.

Most laboratory sources of gamma radiation emit at levels of intensity corresponding to single-photon conditions and Mössbauer experiments are rarely conducted at such great intensities that the detection of two photons would be probable in the transit time spent between source and absorber. Under those conditions, the perception of gamma rays as streams of particles is instinctive. Nevertheless, as elements of electromagnetic radiation they must also be considered as carrier waves of high frequency and analogs of the multiphoton processes which are routine at the molecular level must be possible at the nuclear scale. For years we had maintained that the

key to the excitation of significant levels of multiphoton phenomena lay in the use of small oscillating fields to manipulate the greater ferromagnetic and ferroelectric fields in which the nuclei were immersed. Unfortunately, such materials are almost always magnetostrictive or piezoelectric and the concern had lingered that all coherent phenomena would be overwhelmed and degraded by periodic Doppler shifts produced by vibrations excited by such mechanical efforts. As part of this work we completed the breakthrough experiment[6] in which we dressed nuclear states with photons from the periodic oscillation of magnetization transported by spin waves through a medium unable to propogate vibrations. Large effects were found to confirm our models.

Along the third line of concern to the gamma-ray laser, efforts have been concentrated upon the development of thin film diamond. Diamond has the highest Debye temperature and greatest thermal conductivity of any material. With such a low atomic number there is little absorption of x rays by the electrons and what is stopped does little damage because the waste heat is readily conducted away. Diamond is the ideal medium into which to insert the nuclei to be pumped with single-crystal Be following as a close second. Both will support the Mössbauer effect over as wide a range of aggressive pump conditions as is possible.

In the past few years we have developed a unique means for depositing thin film diamond using a laser plasma source of C^+ ions which are quenched onto a substrate. This seems an ideal environment in which to implant active nuclei, once it becomes known which are the best candidates. The properties and techniques for preparing our laser plasma diamond appear in the literature[7,8,9] and further development and even commercialization for other purposes is being supported by an Austin, Texas company, Research Applications, Inc.

ACHIEVEMENTS

This grant provided for the completion of our efforts to demonstrate that the analogs of quantum electronics exist at the nuclear level. Our understanding is that we did this job so well that a new field of study is condensing as a consequence of our feasibility demonstration. Apparently, the first international workshop on nuclear quantum optics will be organized next year by the Catholic University of Leuven to provide a forum for critical review and discussion.

At the atomic level the studies of phenomena associated with coherently excited states of matter have revolutionized the traditional fields of optics and spectroscopy. Those efforts have benefited greatly from techniques that exchanged the readily available coherence of a radiation field for coherent states of material excitation. In contrast, it had seemed at first that the low intensities of virtually all sources of quanta large enough to be used for the excitation of nuclear states, together with the shortened dephasing times that might characterize the coherent excitation of such levels, would have deterred comparable studies of excited nuclei.

Our early theoretical estimates[10-17] had suggested the opposite; namely that coherent superpositions of states of nuclear excitation might be produced or altered to a readily detectable extent by intense fields of longer wavelength electromagnetic radiation. Rather diverse processes involving such effects were discussed and modeled, including our efforts at quantitatively describing multiphoton absorption, emission, and mixed Raman-type scattering,[10-17] stimulated γ radiation[11-17] and even some particle processes such as induced neutron capture and fission.[13,14,17] A very fertile interdisciplinary area of nuclear quantum electronics[16] appeared to be feasible and we were encouraged by the further theoretical and experimental results of our subsequent work[18-32] under previous ONR contracts. Now, it seems that our enthusiasm has been communicated to a larger community.

In 1978, West and Matthias[33] had reported the first clear case of the excitation of a coherent nuclear state with long-wavelength radiation. They had succeeded in producing at the level of the noise, two radiofrequency sidebands on the 6.1 keV transition of ^{181}Ta in nonmagnetic tungsten. Working with frequencies low enough to be comparable to the Mössbauer width of the transition, at kilowatt levels of input power sidebands finally became perceptible. In 1981, Olariu et al.[14] showed theoretically as a consequence of more general modeling of processes of coherent nuclear excitation[10-13] that a multiphoton model comparable to the Matthias estimate[33] could explain in both magnitude and detail that much stronger development of sidebands in ^{57}Fe experiments. However, the Olariu-Matthias representations had been constructed under the assumption that static magnetization was absent in the Mössbauer absorber and as a result that model was rigorously appropriate only for nuclei embedded in paramagnetic samples. It was one of the first tasks of our group to test the Olariu-Matthias model as an approximation for ferromagnetic ^{57}Fe and, if possible, to resolve experimentally the degree to which results might be contaminated by contributions from magnetostrictive-acoustic effects.

The completion of that task succeeded in confirming the encouraging magnitudes of the sidebands while calling attention to the clear need for a theoretical description more appropriate than the Olariu-Matthias "paramagnetic" model. Subsequently, our group built a greatly extended database and still we found intense sidebands on hyperfine components of the 14.4 keV Mössbauer transition to be maximized under conditions that minimized chances for spurious magnetostrictive effects. Prominent sidebands were observed at only tenths of Watts in input power. Ambiguities in the identification of the order and parentage of particular sidebands were resolved by several changes of frequency. No sidebands were found on forbidden transitions in contrast to the predictions of the simple paramagnetic model.

Another of the tasks under our previous ONR contracts was to construct a multiphoton model for ferromagnetic environments. That description accommodated all phenomena previously reported and also showed that "forbidden sidebands" should not be expected for nuclei embedded in real magnetic materials.

Very significant was the result that unlike the paramagnetic case this new model predicted a dependence of sideband intensity upon power that was drastically different from what would be expected from spurious mechanical vibrations driven by magnetostriction. Gone were the nodes of power at which low-order sidebands would be quenched, a predicted phenomenon receiving no support from observations spanning the past 20 years. Even more significant was that the new multiphoton model of nuclear sidebands was comprehensive enough to suggest new extrapolations and effects not previously apparent. The most exciting was the extensive range over which sidebands might be tuned. This together with indications that it should be possible to drive a large part of the modulated ferromagnetism (that is needed for a large effect) into a different material with other active nuclei seemed to imply the clear feasibility of the swept frequency source for nuclear spectroscopy which was subsequently realized.

Quite independently and from completely different directions,[34-37] concepts were converging upon the same means to excite multiphoton effects at significant levels. From a classical basis a Finnish group developed a model based upon the modulation of the phase of a nuclear state,[34-36] and from scattering theory a Soviet group[37] developed a quantum theory for an unrealistic type of magnetic material. Our own concepts are best characterized as a nuclear analog to the dressed state theory for atoms of Cohen-Tannoudji.[38]

Though completely obscured by different terminology and by the gulf between classical and quantum physics, ours and the Finnish representation are the same. Because of its clarity, the Finnish description is superior since we are able to justify it quantum mechanically.

For cases in which there is no static magnetic field[33] or in which the modulation is parallel to the static field[14] the effect of the time varying component, $H_0 f(t)$ upon a nuclear eigenstate of the nucleus, $\Psi^{(0)}_{\alpha m}$ can be written,

$$\Psi_{\alpha m} = \exp[i\phi_\alpha(t)]\, \Psi^{(0)}_{\alpha m} \quad , \tag{1}$$

where $\phi_\alpha(t)$ is the modulation angle of the phase,

$$\phi_\alpha(t) = m\omega_\alpha \int_0^t f(t')dt' \quad , \tag{2}$$

and the Larmor frequency, ω_α is,

$$\omega_\alpha = \mu_N g_\alpha H_0/\hbar \quad , \tag{3}$$

where μ_N is the nuclear magneton, g_α is the gyromagnetic ratio for the α-th excited or ground state of the nucleus, and m is the magnetic quantum number of the eigenstates.

In principle, the difference in phase modulation between the ground state, g and an excited state, e may be observed during an absorption transition because the Fourier components of $\phi_e(t) - \phi_g(t)$ will be manifest as sidebands. However, since the transition will have a width, Γ associated with the time-dependent decay of the states, unless,

$$\hbar\omega_\alpha \geq \Gamma \quad , \tag{4}$$

the sidebands will be buried in the natural wings of the probing transition.

The Finnish group had derived the factor $\exp[i(\phi_e - \phi_g)]$ and obtained the sidebands but only for this case of linear media. With this restriction coherent effects will inevitably be small. Our conceptual key to orders-of-magnitude of enhancement of phase modulation effects has been reported[20-22] to lie in the use of smaller powers to manipulate the greater magnetic fields arising from the natural correlations of individual spins in ferromagnetic materials. However, the modulation $\partial M/\partial t$ of the magnetization, M of a ferromagnetic material is rarely parallel to either

the applied field H_0 or even to M itself.[39] For such cases of nuclei in ferromagnetic media the modulation angle of Eq. (2) takes a more complex form;[20] and one which causes a mixing of the eigenstates, $\Psi^{(0)}_{\alpha m}$. Nevertheless, the principal parameter is still a Larmor frequency, Ω_α which for magnetic environments becomes,

$$\Omega_\alpha = \mu_N g_\alpha M/\hbar \quad , \tag{5}$$

a value much larger than that found in Eq. (3) for nonmagnetic samples.

The approximate phase angle we developed[21] becomes,

$$\phi_\alpha(t) = \beta_\alpha(\omega_0\Omega_g/\omega^2) \sin\phi_\alpha (\sin\omega t - \omega t\cos\omega t) \quad , \tag{6}$$

where ω is the frequency of reversal of the applied fields, ω_0 is the magnon Larmor frequency, ϕ_α is the average angle between the perturbation and the easy axis of the material,

β_g is 1 and $\beta_e = -3 \mid \omega_e/\omega_g \mid$.

In ferromagnetic foils our expression for the nuclear phase of Eq. (6) replaces the Finnish expression of Eq. (2) for linear materials. Sidebands are computed the same way from the Fourier coefficients of $\exp[i(\phi_e - \phi_g)]$; while the dressed states come from the Fourier expansions of the individual $\exp[i\phi_e]$ and $\exp[i\phi_g]$.

Essentially, the results are the same with the ferromagnetic media scaling as $(\omega_0\Omega_g/\omega^2)$ while the nonmagnetic materials would be limited to the relative size of ω_g/ω. Since $\Omega_g >> \omega_g$ by the ratio of magnetization to free space field strength, and $\omega_0 >> \omega_g$ because of the ratios of nuclear mass to electron mass the effects will be much larger in magnetic media whenever applied frequencies ω are comparable to the static splittings of the nuclear transitions of the order of ω_e and ω_g.

Motivated by the convergence of theories arising in such disparate circumstances we focused considerable attention upon the disturbing impression in the Mössbauer community that an acoustic origin had been "proven" for all sidebands by the benchmark experiment of Chien and Walker[40] in 1976. In that experiment an absorbing foil composed of ferromagnetic and nonmagnetic layers was used to study transport of the causative agent from the ferromagnetic layer into the nonmagnetic region where the sidebands were produced upon Mössbauer transitions of embedded ^{57}Fe nuclei. Very clear evidence showed that the cause did arise in the ferromagnetic Ni layers, producing sidebands in the nonmagnetic stainless steel layers. The most ready explanation at that time was a transport of phonons from one layer to the next with a high acoustic Q. Those experiments were repeated[41] but with extensions which contradicted the classic interpretation of Chien and Walker.[40] In fact, our reexamination showed the original experiment to have been so flawed that any conclusions drawn from it must now be considered unproven.

The propagation of magnetoelastic waves is a complex problem which has been intensively studied[42] since 1958. For many magnetic media the dispersion equation for such waves displays several branches[43,44] which can be individually identified with spin waves, magnetostatic waves or elastic waves. Mixed waves coupling magnons and phonons occur principally when branches intersect, so that the frequencies and wavelengths for both are nearly equal.[42] Away from those values of parameters magnetic and acoustic waves can be separated. That possibility was successfully exploited as a means to propagate only the former to some remote part of a sample where it was desired to magnetically modulate the phases of the states of the nuclei without carrying acoustic noise along to the same place.

The most convenient of the Mössbauer transitions for modulation experiments is the 14.4 keV transition of ^{57}Fe diluted in a thin metal foil. The propagation of magnetic waves in conductive foils presents a special problem because of eddy current losses. Kittel has given an

approximation[42] which would limit the mean free path for a magnon to a few wavelengths for the frequencies of tens of MHz which would be interesting for use with ^{57}Fe. For this reason the preferred choices for the propagation of magnetization in such thin metallic foils are the magnetostatic waves characterized by long wavelengths and high group velocities[43,45] that are quite removed from intersections with acoustic branches. Wavelengths can readily reach the scale of millimeters, and it has been demonstrated that dispersion properties are little affected by raising the temperature of the foil above the Curie point.[46] Thus, if they can be excited at the boundaries, such spin waves should propagate in paramagnetic foils as well as they do in conductive ferromagnetic foils.

Relatively recently, it has been shown[47] how to communicate the large values of magnetization characteristic of ferromagnetic materials into thin foils of paramagnetic media, such as stainless at room temperature, by sandwiching it between ferromagnetic layers. Grünberg demonstrated that at small separations ferromagnetic foils switch coherently so that lines of fringing flux emerging from one continue across to the other. Flux refraction insures that this small normal component is compressed in the separating layer by a factor comparable to its ratio of length to thickness, an aspect ratio of about 10^4 in these experiments. In this way a wave of oscillating magnetization was launched into a stainless tape, enriched in ^{57}Fe so that nuclear phase modulation might be observed at a distance from the source of the disturbance that was greater than the range for the transport of acoustic phonons. In this way an extensive and compelling series of measurements was published that showed the strong degree to which coherent radiation could affect the coupling of the radiation field to states of nuclear excitation.[6]

After this feasibility demonstration all that remained to complete in this direction was to finish a few "loose ends" and to place the work in the context of concepts for gain without inversion. The former led us to the discovery of thermomechanical modulation of gamma

radiation. In the course of exciting sidebands with spin waves, it was observed for the first time that a radiofrequency current flowing through a thin foil would excite sidebands on gamma-ray transition from nuclei in the foil, *but that only sidebands of even order appeared.*

Inspection of Eq. (1) shows that in the most general sense, nuclear phase can be contributed by any interaction energy between the nucleus and its environment. Our first impression was that the Coulomb interaction between the nucleus and the s electrons would contribute a phase for which modulation would have the proper symmetry to suppress sidebands of odd order. It is not unusual for conductions electrons in transition metals to include a proportion of those in s states which mix with the core electron wavefunction. There is a reasonable component of the isomer or chemical shift of nuclear levels from this effect.

Because s electrons have charge distributions that are spherically symmetric, oscillations back and forth across a nucleus would result in a change of interaction energy that is the same for a perturbation to +x or to -x. Then, a mechanism could be conceived in which sinusoidal oscillation of the conduction electrons in the direction of a linear current flow could cause oscillation of the s electrons in the same direction across the nuclear center resulting in an interaction energy containing a rectified sinusoidal term. That term would give only even orders of sidebands in the Fourier expansion of the resulting nuclear phase. Such an effect would point to a very attractive means of manipulating nuclear states by modulating the isomeric shift, but ultimately that did not prove[48] to be the origin of the even sidebands developed by flowing current in the absorbing foil.

Another nuclear phase associated with the position of the nucleus in a medium is,[48]

$$\phi_x(t) = 2\pi x(t)/\lambda \quad . \tag{7}$$

This, however, is the phase of the origin of the γ-ray emission. From our perspective this is a much less interesting quantity as it does not reflect or affect the internal phase of the nuclear state shown in Eq. (6). This phase of Eq. (7) is the one which is produced by vibration or other mechanical effects.

If we assume that the heating associated with the current, i_0 flowing through a localized portion of the foil causes a linear expansion

$$\Delta x = \alpha\lambda_l(\Delta T)/2 \quad , \tag{8}$$

where α is the thermal expansion coefficient and λ_l is the wavelength for an acoustic vibration with velocity v_l; then even order sidebands will be expected because the phase can be written,

$$\phi_x(t) = \frac{2\pi x_0}{\lambda_\gamma} \sin 2\omega t \quad , \tag{9}$$

where

$$x_0 = \frac{\alpha i_0^2 R\lambda_l}{4C\rho V\omega} \quad , \tag{10}$$

and λ_γ is the wavelength of the 14.4 keV γ photon, $C\rho V$ is the heat capacity of the volume and R is the resistance of the foil. Quite clearly a Fourier expansion of the frequencies of a gamma-ray wave emitted with the phase of Eq. (10) will show only sidebands of even order as actually observed.

The reasonable agreement between theory and experiment finally reported[48] strongly suggests that this new type of sidebands has a thermomechanical origin. The likelihood that such a curious phenomena had this trivial origin was supported by the observations that it was very easy to damp them mechanically. Unlike the spin wave sidebands, the phenomenon producing only

even sidebands can be eliminated by contacting the foil to some soft material. In the absence of any indications that there was an unquenched component which might have arisen from a modulated isomer shift, this effect was not investigated further.

Finally, it is a particular pleasure to report that we succeeded in catching Olariu's interest in helping to place the multiphoton sideband work into the context of what is now being termed electromagnetically induced transparency. Appearing in Appendix I is the manuscript "Quantum Interference in the Mössbauer Spectrum of ^{57}Fe Induced by Radiofrequency Magnetic Fields," by S. Olariu, T. W. Sinor, and C. B. Collins. There is it shown that at some radiofrequencies the transition probability for absorption between certain dressed nuclear states becomes zero because of interference between the possible paths for the absorption event. If a single dressed state were initially populated then stimulated emission could occur without the possibility for reabsorption.

This seems to be a substantial victory for efforts trying to demonstrate feasibility of a gamma-ray laser because it shows that gain is possible without inversion at the nuclear level. Of course, the implementation of this effect must wait for an effective way to pump nuclear populations as is being developed along the first line of research described in the Introduction. Therefore, it seems an appropriate time to pause in this line of investigation while efforts are continued to identify the best nuclei to pump. After those critical details are determined and with them the specific nuclear parameters that are pertinent, it would be appropriate to start to apply the principles learned in this work about electromagnetically induced transparency.

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APPENDIX I

Reports of publications appearing during this reporting period.

'Comment on Mössbauer Sidebands from a Single Parent Line",' by C. B. Collins, P. W. Reittinger, and T. W. Sinor, Phys. Rev. B 39, 9655 (1989).

"Large-Scale Effects of the Magnetic Phase Modulation of Recoilless γ Transitions," by T. W. Sinor, P. W. Reittinger, and C. B. Collins, Phys. Rev. Lett. 62, 2547 (1989).

"Mössbauer Isomer Shift Measurements without Mechanical Tuning," by T. W. Sinor, P. W. Reittinger, and C. B. Collins, Rev. Sci. Instrum. 60, 3258 (1989).

"Thermomechanical Frequency Modulation of γ Radiation," T. W. Sinor, O. Y. Nabas, J. D. Standifird, and C. B. Collins, Phys. Rev. Lett. 66, 1934 (1991).

"Quantum Interference in the Mössbauer Spectrum of ^{57}Fe Induced by Radiofrequency Magnetic Fields," by S. Olariu, T. W. Sinor, and C. B. Collins.

Comment on "Mössbauer sidebands from a single parent line"

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Foils composed of alternating layers of ferromagnetic and nonmagnetic materials immersed in magnetic fields oscillating at radio frequencies display sidebands on Mössbauer transitions from the nuclei contained in the nonmagnetic regions. Attributed by Chien and Walker [Phys. Rev. B 13, 1876 (1976)] to the transfer into the nonmagnetic layer of acoustic phonons excited by magnetostriction in the ferromagnetic layers, this accepted cause of such effects is challenged by new data resulting from a reexamination and extension of that classical experiment.

The paper of Chien and Walker[1] was of such critical importance that it warrants comment over a decade later. Generally perceived as reporting an unarguable proof of a certain basic proposition, it has now been found to have rested upon a demonstrably false assumption. A reexamination of the original experiment shows it to have been so flawed that any conclusions drawn from it must now be considered unproven.

The point of inception had been the original proposal of Mitin[2,3] that Mössbauer transitions could be excited as part of a multiphoton process in nuclei immersed in intense radio-frequency (rf) fields. In those cases the Mössbauer spectrum was expected to show additional sum and difference frequency lines displaced from the normal lines by integral multiples of the perturbing frequency. In appearance such multiphoton spectra are expected to resemble the transmission spectra which Ruby and Bolef[4] obtained by imposing periodic Doppler shifts of purely mechanical origin upon the Mössbauer source. This unfortunate similarity in appearance between phenomena arising from such different origins provided the basis for years of critical controversy seemingly resolved by the work of Chien and Walker.[1] The purpose of this comment is to report new data from a repetition and extension of the Chien and Walker experiment that shows their conclusions to be unjustified. Without the force of conviction conveyed by their work, the controversy must be reopened to further investigation.

The earliest experiment in radio-frequency sideband production, reported by Perlow[5] in 1968, focused upon the components of the 14.4-keV transition in ^{57}Fe. Several ^{57}Co sources diffused into ferromagnetic hosts were immersed into intense magnetic fields oscillating at radio frequencies. Those results were attributed[5] to successive jumps in the hyperfine energies of the radiating states that had been caused by rotations of the local direction of the magnetization at the nucleus. Those jumps were assumed to occur at the random times when domain walls passed, but with an average periodicity which was treated as a fitted parameter. Such a treatment could be termed a magnetodynamic model of sideband development which anticipated the results of the more complex propagation of the rotations of the magnetization in foils and tapes that was subsequently developed.[6,7] It generally conformed to the Mitin hypothesis for multiphoton transitions. Two of the three groups who initially documented this phenomena favored the magnetodynamic explanation which required no mechanical action[5,8,9] while the other group began to develop an alternative based entirely upon magnetostriction.[10,11] Most of the actual experiments had used ferromagnetic hosts to enhance the applied magnetic fields, and such materials are almost invariably magnetostrictive. In the model finally synthesized, periodic Doppler shifts were assumed to be driven by acoustic phonons which were excited by magnetostriction along the greatest dimensions of the material and scattered onto the axis connecting source and absorber. To be effective, this mechanism required the sample to have a large acoustic Q so that displacements of the active nuclei could build to significant values.

Despite the accretion over the years of a large body of phenomenology presumed to described rf sidebands on Mössbauer transitions, the magnetostrictive-acoustic theory never quantitatively predicted the amplitudes of the sidebands as functions of either applied power or frequency. However, the magnetodynamic models of that time fared no better, and attention turned to "proving" a magnetostrictive origin by distressing the alternative explanations.[12] The obvious difficulty with proving a theory by distressing the alternatives is that those other explanations may not have reached comparable levels of maturation. The magnetodynamic models of the late 1960's were relatively easy to destroy.[12] However, the recent successes of ferromagnetodynamics[6,7] show the early models[5] of

sideband formation to have been inspired, but inadequate approximations. Those models simply did not embody the level of sophistication necessary to describe the complex switching behavior of magnetization in ferromagnetic foils subjected to various combinations of static and oscillating fields in those geometries employed.

More recent experiments[13,14] have shown that the applications of such oscillating magnetic fields to Mössbauer nuclei embedded in nonmagnetic hosts do produce radio-frequency sidebands by directly modulating the phases of the nuclear states involved in the transitions. However, amplitudes were rather small in those experiments because the driving forces depended only upon the value of applied field $\mu_0 H$. In 1984, we extended such approaches further by deriving the phase modulation of a nuclear state in a magnetic material.[15] In this case driving forces were proportional to the magnetization $\mu_0 M$ and effects were found to be large.[15-17] It appears that many prior results attributed exclusively to acoustic effects driven by magnetostriction could have also benefited from an unrecognized contribution from direct phase modulations of the nuclear states involved.

From a current perspective it is the experiment reported by Chien and Walker[1] that forms the bulwark of the magnetostrictive-acoustic explanation of Mössbauer sidebands. In that experiment an absorbing foil composed of ferromagnetic and nonmagnetic layers was used to study transport of the causitive agent from the ferromagnetic layer into the nonmagnetic region where the sidebands were produced upon Mössbauer transitions of embedded ^{57}Fe nuclei. Very clear evidence showed that the cause did arise in the ferromagnetic Ni layers, producing sidebands in the nonmagnetic stainless-steel layers. The most ready explanation at that time was a transport of phonons from one layer to the next with a high acoustic Q. Those experiments were repeated in the work reported here, but with extensions which contradict the classic interpretation of Chien and Walker.[1]

Although not unique for all sidebands in a spectrum,[1] the idea of a modulation index m as a measure of the strength of the development of the sidebands offers practical convenience for descriptions. For a magnetostrictive origin,[1]

$$m = x_0/\lambda , \tag{1}$$

where x_0 is the amplitude of the periodic displacement of the nuclei and $\lambda = 0.137$ Å for the 14.4 keV line of ^{57}Fe. In the corresponding magnetodynamic model,[15]

$$m = bH , \tag{2}$$

where H is the applied magnetic field and b provides proportionality between M_s, the saturation magnetization of the medium, and H. For relatively small m, the ratio of the magnitude of the first order sidebands to the intensity in the original parent line is proportional to m^2, which in turn is proportional to P, the applied radio-frequency power.

One of the most compelling results presented by Chien and Walker[1] was a demonstration supposed to show the enhancement of m^2 afforded by tighter acoustic coupling of the layers. They found that electroplating Ni upon a stainless-steel foil produced much higher values of m^2 in absorption experiments than could be obtained by gluing a Ni foil to the stainless foil. They attributed the difference to the obviously poorer acoustic properties of the glue. However, as part of this report we observe that their stainless-steel foil was electroplated on *both* sides with Ni while the epoxied bond was used to join a *single* Ni foil to one side of the stainless absorber. While the m defined by Eq. (1) for a single foil could not be additive if produced in different magnetostrictive layers, in principle the H upon which m depends in Eq. (2) could add coherently. Two sources of m arising from distinctly separate sources could give a resulting modulation of $4m^2$ in a magnetodynamic model. Chien and Walker failed to recognize[1] that even in the magnetostrictive model two sources of m generated in the two electroplated layers should give a modulation index of $2m^2$ in the absorber foil. Instead, they attributed the increased sideband intensity developed by the two plated sources in comparison to the one glued source only to the advantage they assumed for a plated contact over a glued interface. They reported no comparison of the effects of gluing or plating the *same number of ferromagnetic layers to the absorber foil.* Reported here is a repetition of the Chien and Walker experiment which showed that the effect of two foils varied from two to four times that produced by a single foil joined in the same fashion, depending upon the static magnetic bias applied.

In our experiment the absorber was a 2.5 μm paramagnetic stainless-steel (SS) foil with 90.6% enrichment of ^{57}Fe. For the nonabsorbing ferromagnetic drivers, 2.5 μm Ni foils were used, all of which were cut from a single sheet of polycrystalline Ni. The stainless-steel absorber was sandwiched between two Ni foils and held in rigid contact by mounting the foils between glass cover slides of 100 μm thickness. A conventional Mössbauer spectrometer, modified for rf experiments (Fig. 1), utilized a 25-mCi source in a Rh matrix to obtain the ^{57}Fe absorption spectra. The 14.4-keV gamma rays were detected with a Kr gas filled proportional counter biased with 1.8 kV.

A 25 MHz rf magnetic field was applied by mounting the foils in the cylindrical inducation coil of an L-C tank circuit. In obtaining data for a direct comparison between the effect of one Ni driver versus two, the product of the applied rf power P and the electrical Q of the circuit containing the rf induction coil was maintained at constant values. Elementary analysis shows that if PQ is constant the rf current in the coil of such a circuit is also constant and hence the two absorber arrangements are subjected to applied fields of the same intensity H. The results of the first experiment verified the linearity of the first-order sideband amplitudes at 25 MHz for SS with two Ni drivers with PQ products of 75, 150, and 300 W as shown in Fig. 2. The spectra are scaled so that the intensity of the central Mössbauer absorption peak of ^{57}Fe in SS is held constant in order to make direct comparisons of the sideband amplitudes.

Having established the linearity of the first-order sidebands in the Ni-SS-Ni sandwich, one of the Ni drivers was removed and the experiment was repeated with the same PQ products as before. Figure 3 shows a comparison of the sideband amplitude for two Ni drivers versus

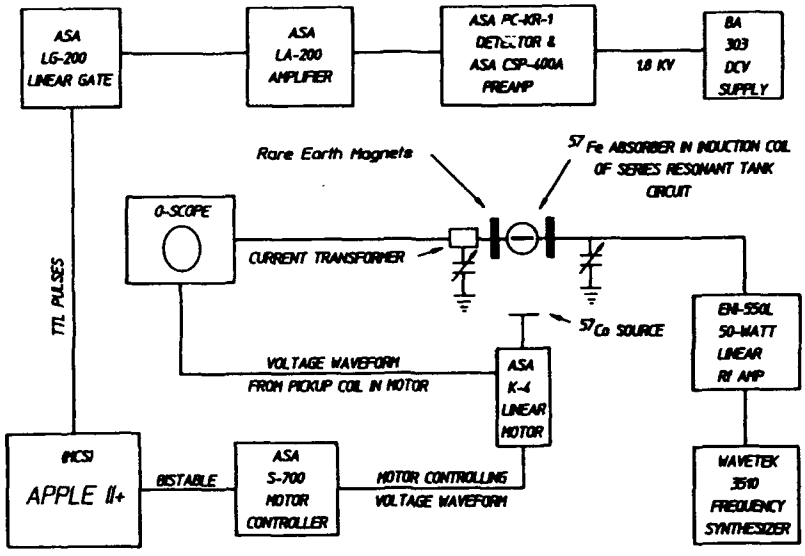

FIG. 1. Schematic drawing of the experimental arrangement used.

one; in this configuration two Ni drivers give twice the effect of one driver foil.

In the next experiments a comparison was made between the effect of one source of excitation with that from two sources when both were biased with a static magnetic field. Rare-earth element magnets were placed about the induction coil such that the static magnetic field was mutually orthogonal to the rf magnetic field and to the direction of gamma-ray propagation.

The linearity of the sideband amplitudes at 25 MHz as a function of PQ was again established as shown in Fig. 4 to insure that the introduction of the static magnetic field did not introduce any nonlinearities into the system. The scale thus established was used to measure the decrease in the sideband amplitude when one of the sources of excitation was removed from this biased sandwich. As is clearly shown in Fig. 5, the sideband amplitudes obtained with two driver foils are four times the amplitudes obtained with one driver foil. Therefore, with the application of a

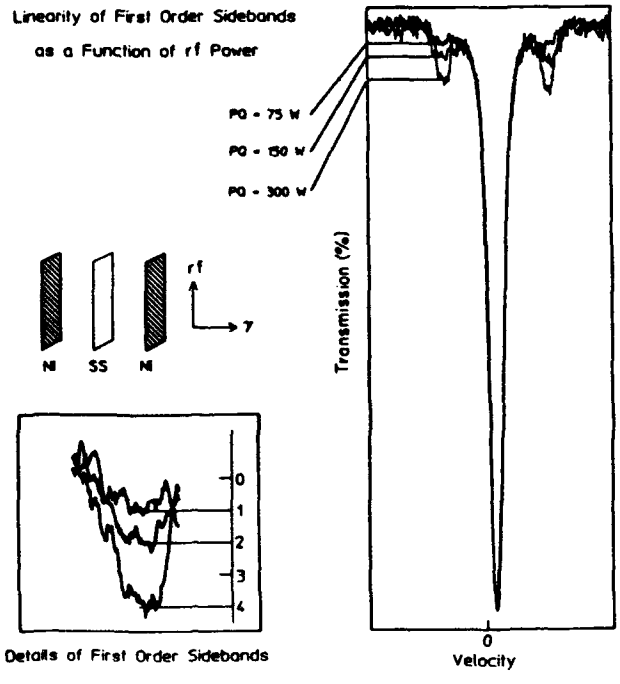

FIG. 2. Experimental verification of the linearity of the first-order sidebands at 25 MHz as a function of the applied rf power. The product of the applied rf power P and the quality factor Q of the circuit are used to insure reproducibility of the rf field strengths.

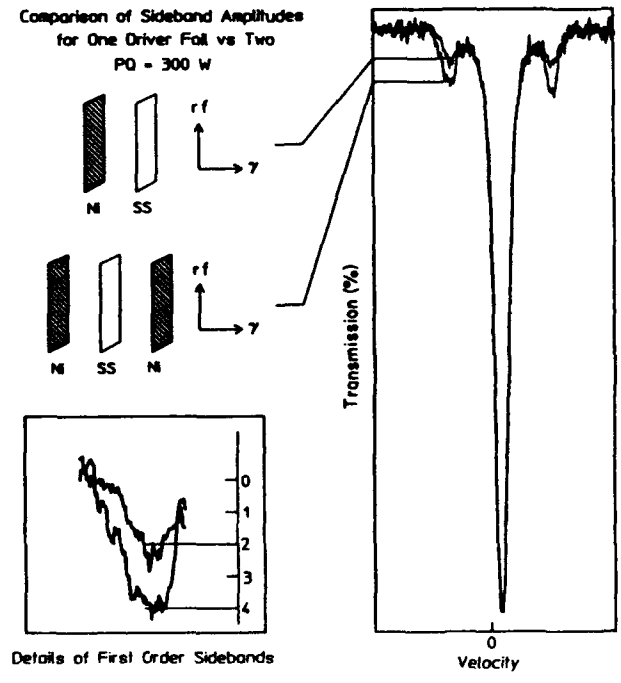

FIG. 3. Comparison of first-order sideband amplitudes for one Ni driver foil vs two at 25 MHz with a PQ product of 300 W.

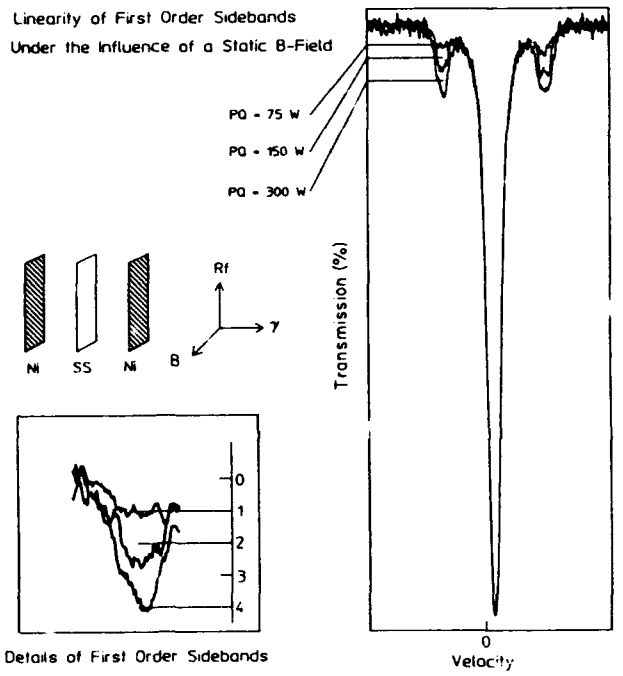

FIG. 4. Establishment of linearity of the first-order sidebands at 25 MHz with $PQ = 75$, 150, and 300 W when the foils are biased with a static B field.

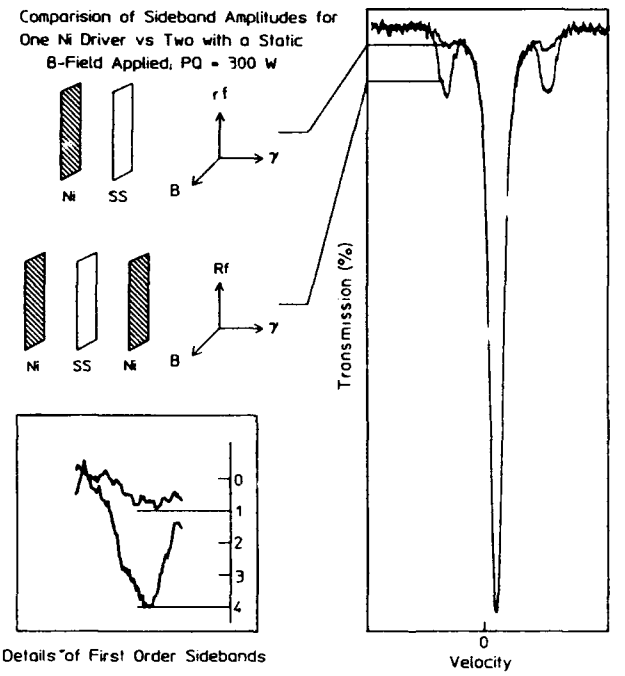

FIG. 5. Comparison of sideband amplitudes for one driver foil vs two when both are biased by a static B field with $PQ = 300$ W. Here two foils give four times the effect of one thus giving a modulation index of $4m^2$.

static B field, two sources of excitation give four times the effect.

The results of this reexamination of the Chien and Walker experiment support only the first conclusion reached in that original work, namely that the causitive agent of rf sidebands can be produced in a ferromagnetic layer and then transported into a nonmagnetic layer. Their other conclusion is completely refuted by this demonstration because the effects they attributed to the type of coupling between layers most probably resulted from the relative numbers of magnetic and nonmagnetic layers.

These new results go beyond the propositions tested by Chien and Walker[1] and display behaviors completely inconsistent with the traditional magnetostrictive-acoustic origin of Mössbauer sidebands. In experiments such as these, acoustic phonons are the bosons associated with vector fields driven by tensor forces, *not vector forces.* Without invoking stimulated emission, we can conceive of no way in which tensor sources which are physically separated can produce coherent vector fields in a space between them, even if they are temporally synchronized. As the fields increase the magnetostrictive foils will become stressed along parallel axes which are displaced by the thickness of the stainless layer between them. There is no mechanism to produce a displacement vector in a particular transverse direction as a consequence of the resulting strains in the Ni foils. Only a small scale bulging or buckling of each Ni foil is to be expected and this is usually described as the scattering of phonons at right angles to the source. Without the stimulated emission of such phonons, there is no way to insure that one foil buckles toward the stainless layer at a particular point while the other buckles away.

The stimulated emission of phonons that would be necessary to produce coherent additions of the displacements arising from the different sources would imply the existence of a threshold of power, above which two modulation indices of m would give an effect of $4m^2$ and below which only $2m^2$. No such threshold was suggested by data similar to that of Fig. 5 which was obtained over an adequate range of powers.

In view of the growing number of successes of the model for the direct modulation of the phases of the nuclear states and these new results which question the validity of the conclusions of the Chien and Walker[1] experiment, it would appear that the controversy over the origin of Mössbauer sidebands must be reopened.

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Large-Scale Effects of the Magnetic Phase Modulation of Recoilless γ Transitions

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The excitation of coherent transients in Mössbauer spectra has been previously limited by the high powers required to modulate the nuclear phases. Reported here is an orders-of-magnitude increase in the efficiency through which such phenomena can be produced. Magnetic modulation of the quantum phases of ^{57}Fe nuclei in paramagnetic media has been produced by spin waves of large amplitude transported from ferromagnetic sources.

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A recent Letter[1] reported variations in recoilless γ-ray spectra produced by directly modulating the interaction energies arising from the couplings of the nuclear magnetic moments to the hyperfine fields. Described in terms of the phase modulation of the nuclear states involved in the transition, those results represent important realizations of some of the general possibilities for developing nuclear analogs[2,3] of coherent transient effects studied in quantum optics.

An example is found in the frequency domain in the excitation of sidebands on γ-ray transitions, and precursive work[4-6] had treated that problem in a manner equivalent to an application of the phase modulation formalism.[3] For cases in which there is no static magnetic field[5] or in which the modulation is parallel to the static field,[6] the effect of the time-varying component $H_0 f(t)$ upon an eigenstate of the nucleus, $\Psi_{a,m}^{(0)}$, can be written

$$\Psi_{a,m} = e^{-i\phi_a(t)}\Psi_{a,m}^{(0)}, \tag{1}$$

where $\phi_a(t)$ is the modulation angle of the phase,

$$\phi_a(t) = m\omega_a \int_0^t f(t')dt', \tag{2}$$

and the Larmor frequency ω_a is

$$\omega_a = \mu_N g_a H_0/\hbar, \tag{3}$$

where μ_N is the nuclear magneton, g_a is the gyromagnetic ratio for the ath excited or ground state of the nucleus, and m is the magnetic quantum number of the eigenstates.

In principle, the difference in phase modulation between the ground state, g, and an excited state, e, may be observed during an absorption transition because the Fourier components of $\phi_e(t) - \phi_g(t)$ will be manifested as sidebands. However, since the transition will have a width Γ associated with the time-dependent decay of the states, unless,

$$\hbar\omega_a \geq \Gamma, \tag{4}$$

the sidebands will be buried in the natural wings of the probing transition.

Only the ingeneous use of the ultranarrow, 93-keV line of ^{67}Zn permitted the quantitative study of the coherent phase modulation effects recently described.[1] That benchmark achievement required field amplitudes reaching 13.4 mT for sinusoidal modulation at applied frequencies ω up to 10 kHz and effects were reported[1] to scale as $(H_0/\omega)^2$.

The conceptual key to orders-of-magnitude enhancement of phase modulation effects has been reported[7-9] to lie in the use of smaller powers to manipulate the greater magnetic fields arising from the natural correlations of individual spins in ferromagnetic materials. However, the modulation $\partial M/\partial t$ of the magnetization M of a ferromagnetic material is rarely parallel to either the applied field H_0 or even to M itself.[10] For such cases of nuclei in ferromagnetic media, the modulation angle of Eq. (2) takes a more complex form;[7] and one which causes a mixing of the eigenstates $\Psi_{a,m}^{(0)}$. Nevertheless, the principal parameter is still a Larmor frequency Ω_a which for magnetic environments becomes

$$\Omega_a = \mu_N g_a M/\hbar, \tag{5}$$

a value much larger than that found in Eq. (3) for nonmagnetic samples. Unfortunately, magnetic materials are almost invariably magnetostrictive and the concern has lingered that even the enhanced effects of phase modulation might always be overwhelmed by the periodic Doppler shifts produced by vibration in the lattices excited by magnetostriction.

The propagation of magnetoelastic waves is a complex problem which has been intensively studied since 1958.[11] For many magnetic media the dispersion equation for such waves displays several branches[12,13] which can be individually identified with spin waves, magnetostatic waves, or elastic waves. Mixed waves coupling magnons and phonons occur principally when branches intersect, so that the frequencies and wavelengths for both are nearly equal.[11] Away from those values of parameters magnetic and acoustic waves can be separated. In principle this offers a means to propagate only the former to some remote part of a sample where it is desired to magnetically modulate the phases of the states of the nuclei without carrying acoustic noise along to the same place.

The most convenient of the Mössbauer transitions for modulation experiments is the 14.4-keV transition of

^{57}Fe diluted in a thin metal foil. The propagation of magnetic waves in conductive foils presents a special problem because of eddy-current losses; Kittel has given an approximation[11] which would limit the mean free path for a magnon to a few wavelengths for the frequencies of tens of MHz which would be interesting for use with ^{57}Fe. For this reason the preferred choices for the propagation of magnetization in such thin metallic foils are the magnetostatic waves characterized by long wavelengths and high group velocities[12,14] that are quite removed from intersections with acoustic branches. Wavelengths can readily reach the scale of millimeters, and it has been demonstrated that dispersion properties are little affected by raising the temperature of the foil above the Curie point.[15] Thus, if they can be excited at the boundaries, such spin waves should propagate in paramagnetic foils as well as they do in conductive ferromagnetic foils.

A stainless-steel foil 10 mm×20 mm×2.5 μm was used in these experiments. It was rolled from a nonmagnetic alloy, 310, which was expected[16] to have a susceptibility of about 4000×10^{-6} cgs. When enriched in ^{57}Fe, it displayed a single absorption line at 14.4 keV. Relatively recently, it has been shown[17] how to communicate the large values of magnetization characteristic of ferromagnetic materials into thin foils of paramagnetic media, such as stainless steel at room temperature, by sandwiching it between ferromagnetic layers. Grünberg demonstrated that at small separations ferromagnetic foils switch coherently so that lines of fringing flux emerging from one continue across to the other. Flux refraction insures that this small normal component is compressed in the separating layer by a factor comparable to its ratio of length to thickness, an aspect ratio of about 10^4 in these experiments. In this way a wave of oscillating magnetization was launched into a stainless-steel tape, enriched in ^{57}Fe so that nuclear phase modulation might be observed at a distance from the source of the disturbance that was greater than the range for the transport of acoustic phonons.

This experiment employed a conventional Mössbauer spectrometer with a 2 mCi ^{57}Co source in a Pd matrix to obtain the ^{57}Fe resonance spectra in a transmission geometry. To detect the 14.4-keV γ rays we used a high-resolution EG&G high-purity Ge detector. A Wavetek Model 3000 signal generator and a 50-dB ENI-325LA linear radio-frequency (rf) amplifier provided the oscillating magnetic field.

To calibrate the contributions from ultrasonic and from phase modulation effects two independent techniques of sideband generation were used. As a basis for comparison, ultrasonic sidebands were excited on the unsplit absorption line of ^{57}Fe nuclei in the same stainless-steel foil by sinusoidal vibrations injected with two different 25-MHz piezoelectric transducers. One was X cut and the other AT cut in order to inject the widest possible variety of phonons of vibration for transmission to the point of observation in the geometry of Fig. 1(a).

As shown in the lower panel of Fig. 1(b) an 8×10 mm^2 section of the foil was acoustically bonded to each of the transducers used in this experiment. The remaining length was gently curved, forming a 90° angle with the plane of the transducer. Mechanical support was provided for the remote section of foil by mounting it between glass cover slides which were then fastened to the transducer cell.

In the usual geometry for a transmission experiment arranged to sample excitation at the source, a convenient level of input power of 0.06 W to the X-cut crystal produced the reference level of sideband development seen

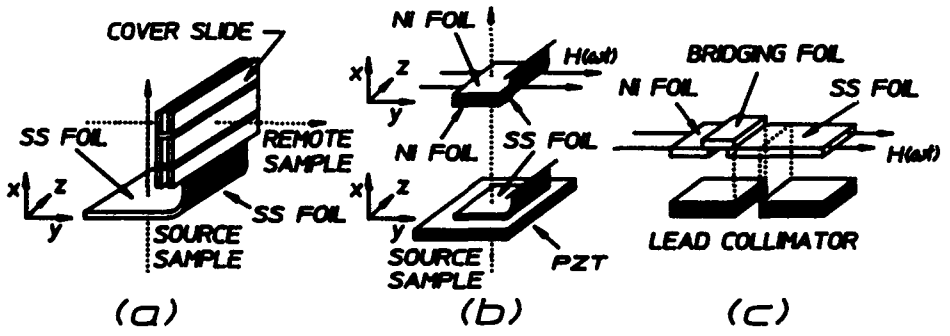

FIG. 1. Schematic representation of the mounting arrangements used in the excitation of sidebands on the Mössbauer absorption line of ^{57}Fe in the 2.5-μm-thick stainless-steel (SS) foil. (a) The optical path from γ source to detector is shown by the dotted arrow for the orthogonal x and y directions arranged to sample sideband development at the source of the phenomenon and at a remote point, respectively, as marked. Excitation is injected into the horizontal face of the SS foil and the vertical face is mechanically stabilized by the 100-μm-thick glass cover slides sandwiching the absorber foil. (b) Upper: spin waves injected by the periodic oscillation of the Ni foils pressed onto the absorber foil by additional cover slides not shown and excited by the magnetic field in an inductor containing the Ni foils. The vertical section protrudes from the coil between windings. Lower: vibrational excitation injected by the X-cut quartz-crystal transducer connected to a radio-frequency oscillator. (c) Geometry used in the experiments designed to bridge a break in the conductive path with 25-μm-thick foils of various materials.

in Fig. 2 in which the fourth order contained 34% of the intensity remaining in the parent transition. Nevertheless, the effect of phonons transported in the foil about 1 cm around a bend of 90° could not be detected even with a tenfold increase in power above the reference level. Nor could they be observed at the remote sampling point when excitation was provided by the *AT*-cut transducer which required a larger input power of 2.9 W in order to obtain the same reference level of sideband development at the source position. The presence of a high level of phonon excitation *in the stainless-steel foil at the source end* was clearly demonstrated by the strong development of the sidebands seen there, but evidently those phonons could not propagate to the remote point of observation. If there had been any low-loss modes for the transport of purely elastic waves in the stainless-steel foil, the population of phonons already in the foil at the source end should have coupled into them. However, the absence of phonon transport actually observed seems consistent with the difficulties expected in propagating transverse vibrations through a medium which is very thin in comparison to a wavelength and one in which the local acoustic Q is expected to be high.[18]

In the experimental arrangement used to launch spin waves, the piezoelectric transducer was replaced with a pair of 2.5-μm foils of ferromagnetic Ni (8×10 mm^2) which were periodically magnetized as shown in the upper panel of Fig. 1(b). The foils (Ni-SS-Ni) were held in rigid contact by sandwiching them between glass cover slides at the source end. Only the source end of the experiment was changed and the protruding section of the stainless-steel foil was again bent at an angle of 90° and enclosed between glass cover slides in the same way as before. Mechanical rigidity was provided to the absorber assembly by a plastic frame.

An rf magnetic field of 0.07 mT was applied to the absorber via a flattened induction coil of a tuned LC circuit. During observation in the x direction at the source end the rf power level was adjusted to give about the same reference level of sideband development as had been generated with the piezoelectric transducer. This insured the same ambient level of phonons in the foil at the source end and thus provided for a direct comparison of the phase modulation effects propagated by magnetostatic waves with the null level of acoustic noise that had reached the same point of observation during calibration.

The spectra obtained in the samples of the source and remote points in the geometry of Fig. 1(b) are shown in Fig. 3. At the remote point for this same level of input power, first-order sidebands containing about 22% of the intensity of the parent line developed. This represents a level 10 times the threshold for detection and hence 100 times any component contributed by acoustic phonons as determined from the data of Fig. 2. It seems that these experiments have shown that an oscillating alignment of spins can be propagated through a paramagnetic foil better than mechanical vibrations can be transported under the same conditions.

Moreover, such effects of spin waves were not sensitive to mechanical pressures or spurious damping and the complete disassembly, substitution of foils, and reassembly served to reproduce closely the levels of excitation seen in Fig. 3. Only the thickness and length of the foil, together with the level of input power, were found to be variables that significantly affected the result.

Finally, to confirm the magnetic nature of the transport of the cause of the sidebands, the Ni foil generating the spin waves was separated from the SS foil by a gap which was then bridged as shown in Fig. 1(c) by foils having various magnetic and acoustic properties. To insure reliable and reproducible contact of the foils the layers were pressed between two thin glass plates. As expected, these plates alone were unable to communicate the spin waves from the Ni to the SS and no sidebands could be produced without a bridging foil.

All of the bridging foils were of a uniform size and thickness, being 4 mm by 10 mm by 25 μm, respectively. They differed only in acoustic and magnetic properties.

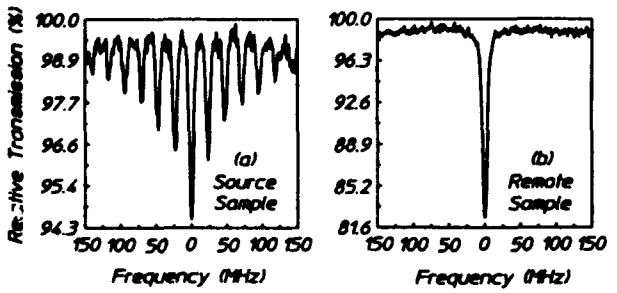

FIG. 2. Absorption spectra of ^{57}Fe in stainless steel showing sidebands developed by phonons present in the sample foil. (a) Reference spectrum observed in the x direction at the source at a level of excitation of 0.06 W in the piezoelectric crystal. (b) Spectrum observed in the y direction at the remote point shown in Fig. 1(a) for an excitation level of 0.7 W. Only the unsplit parent line is seen.

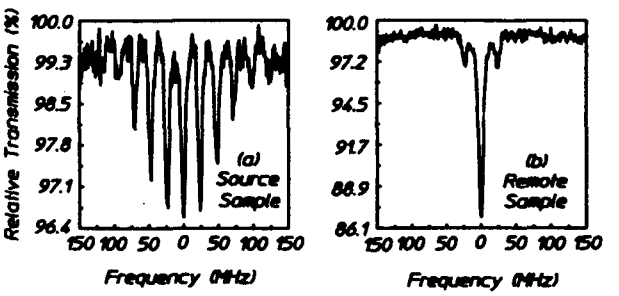

FIG. 3. Absorption spectra of ^{57}Fe in stainless steel showing sidebands resulting from the modulation of nuclear phases by spin waves present in the sample foil. (a) Reference level of excitation observed at the source in the x direction shown in Fig. 1(b) at a field amplitude of 0.07 mT. (b) Sidebands observed in the y direction at the remote point for the same driving field of 0.07 mT.

TABLE I. Summary of experimental results for the rf bridging experiments.

Bridging material	Susceptibility χ (10^{-6} cgs)	Acoustics impedance $Z=\rho c$ (10^5 g/cm^{2}sec)	Effect transported
Aluminum	+16.5	17.33	Yes
Copper	−5.46	44.74	No
Silver	−19.5	37.96	No
310-SS	+4000	45.4	Yes
Tin	−37.0	24.24	No
Titanium	+153	27.32	Yes

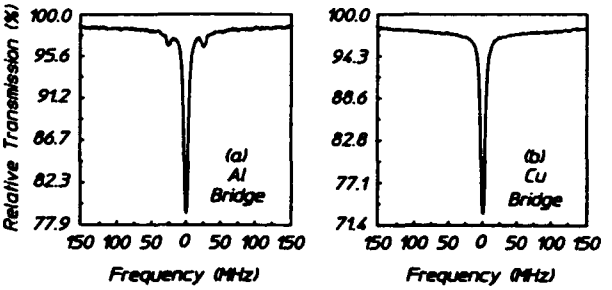

FIG. 4. Absorption spectra of ^{57}Fe in stainless steel in the geometry of Fig. 1(c) for a certain constant level of radio-frequency input. (a) Sidebands developed with an Al bridge. (b) Null effect obtained with a Cu bridge.

As shown in Table I, the transport of the spn waves correlated completely with the susceptibility of the bridge and showed no dependence upon the matchings of acoustic impedances.

For example, consider the case for Al which has a magnetic susceptibility of $+16.5\times10^{-6}$ cgs but which offers a very poor matching of its acoustic impedance to that of either Ni or SS. With an Al bridge there was a strong transport of the cause of the sidebands as seen in Fig. 4(a). In contrast Cu which is diamagnetic, as indicated by its negative susceptibility, should have been unable to propagate spin waves across the gap, despite its very close match of acoustic properties to both Ni and SS. Figure 4(b) shows that sidebands did not develop with the Cu bridge.

While it is expected that the transport of purely acoustic waves should be quite sensitive to interface contacts and damping effects, the spin waves propagated in these experiments were not found to be affected by the contact pressure or by the mechanical mounting. The area of contact and length of the bridge were important parameters but the pressure of the cover glasses and placement of the supports were not important. The data of Fig. 4(a) were routinely reproduced to within the noise level even after disassembly and rearrangement of the mechanical parts.

In these experiments the driving amplitude H_0 in the Ni was around 0.07 mT while the frequency was 23.74 MHz. Without the excitation of spin waves the effect should have been smaller by at least the ratios of the scaling parameters, $(B_0/\omega)^2$, a factor of 10^{11} for this case. The great enhancement in the effects of coherent modulation which spin waves produce makes possible the examination of many other coherent phenomena predicted at practical ranges of tuning and accessible levels of input powers.

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Mössbauer isomer shift measurements without mechanical tuning

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A technique is described which demonstrates how a frequency modulation spectrometer (FMS) can be used to measure the isomer shift of ferromagnetic absorbers without mechanical tuning. As an example, the isomer shift of an iron sample relative to a ^{57}Co source in a Pd matrix was measured and found to be $-(0.1869 \pm 0.003)$ mm/s compared to the literature value of $-(0.185 \pm 0.02)$ mm/s.

INTRODUCTION

The high Q of a Mössbauer resonance, approximately 10^{12} for ^{57}Fe, provides photons of well-defined energy in the keV range that are useful for probing phenomena involving very small changes of energies. In this way Mössbauer spectroscopy can provide a highly detailed account of the way in which the nucleus interacts with its environment. It is only through the existence of the Mössbauer effect that a practical source of narrow line radiation can be produced at these wavelengths, and with such a source very small changes in frequency can be measured.

With Mössbauer techniques there is sufficient frequency resolution that spectral changes produced by magnetic perturbations to the nucleus can be observed in the short wavelength limit where quantum effects are dominant. In particular the Mössbauer effect is ideally suited to the study of the interaction of the nucleus with a rapidly oscillating magnetic field. Mitin[1,2] was the first to propose that Mössbauer transitions could be excited as part of a multiphoton interaction for nuclei immersed in intense radio-frequency fields. The earliest experiments investigating the influence of radio-frequency magnetic fields on Mössbauer transitions was reported by Perlow[3] in 1968. In his experiment, Perlow subjected several ^{57}Co sources of the 14.4-keV transition of ^{57}Fe to intense magnetic fields oscillating at rf frequencies and was able to demonstrate the destruction of the Mössbauer hyperfine pattern by the action of the rf field. Later researchers[4] soon found that when a ferromagnetic iron absorber was subjected to long wavelength photons of an alternating magnetic field with frequency Ω (MHz), the Mössbauer spectrum contained additional absorption lines, known as rf sidebands, at frequencies $\omega_i \pm n\Omega$ where ω is the frequency of the parent transition and $n = 1,2,...$, but these were shown to be artifacts produced by spurious acoustic noise generated in the absorber. True sum and difference frequency lines were not found in Mössbauer spectra until much later.[5-10] The origin of these tunable sidebands in multiphoton or nuclear phase modulation effects was established even more recently.[11-14]

The frequency dependence of these rf sidebands provides the basis for development of a high resolution gamma-ray spectrometer which operates by modulating the cross section for the gamma-ray absorption. A prototype of this Frequency Modulation Spectrometer (FMS) was first described[10] by our laboratory in 1985 with subsequent refinements[15] reported in 1988.

A FMS spectrum of ^{57}Fe provides a direct measurement of rf sideband positions and intensities without interference from the parent transitions. It does this by scanning the frequency of the probing sidebands[15] as opposed to the shifting of the transition energy conventionally employed. In such a spectrum one can extrapolate information about the transitions between Zeeman split energy levels (parent transitions) and the nuclear isomer shift between the source and absorber. In particular, it provides a technique for accurately measuring the isomer shift directly in terms of frequency; a quantity which can be measured more precisely than the customary Doppler velocity.

The heart of the Frequency Modulation Spectrometer is an Apple II + computer which serves as a Multichannel Scaler (MCS) and an IEEE-488 GPIB interface as shown in Fig. 1. The GPIB enables the spectrometer to sweep continuously through the frequencies of the rf magnetic field which is controlled by a Wavetek frequency synthesizer. A Mössbauer linear motor allows the frequency of the incident gamma rays to be biased by a constant Doppler shift, if desired. The spectrometer currently has an instrumental resolution of 100 Hz and a frequency range of 1 MHz–1 GHz with a stability of 0.1 Hz/s when used with a stationary source.

In Fig. 2(a) a conventional Mössbauer spectrum of ^{57}Fe showing the six allowed magnetic transitions is shown without external perturbation. Component lines are labeled 1 through 6 with the transition of lowest energy identified as number 1. In the spectrum of the same absorber subject to the excitation of rf sidebands as shown in Fig. 2(b), the sidebands have been labeled using the following nomenclature. The first digit corresponds to the order of the sideband n, with the second number corresponding to its particular parent transition, p. The sign of n indicates whether the sideband appears at a higher ($+n$) or lower ($-n$) energy than its parent transition. For example, $-1;5$ denotes the first order sideband displaced to lower energies from parent transition 5.

The parent transitions 1 and 6 of Fig. 2(a) are symmetric about the centroid of the resonance pattern and are separated by 123.68 MHz. The application of a 61.84 MHz alternating magnetic field to the ^{57}Fe absorber produces $+1;1$ and $-1;6$ sidebands which overlap at the center of the hyperfine structure of the Mössbauer spectrum. The energies of

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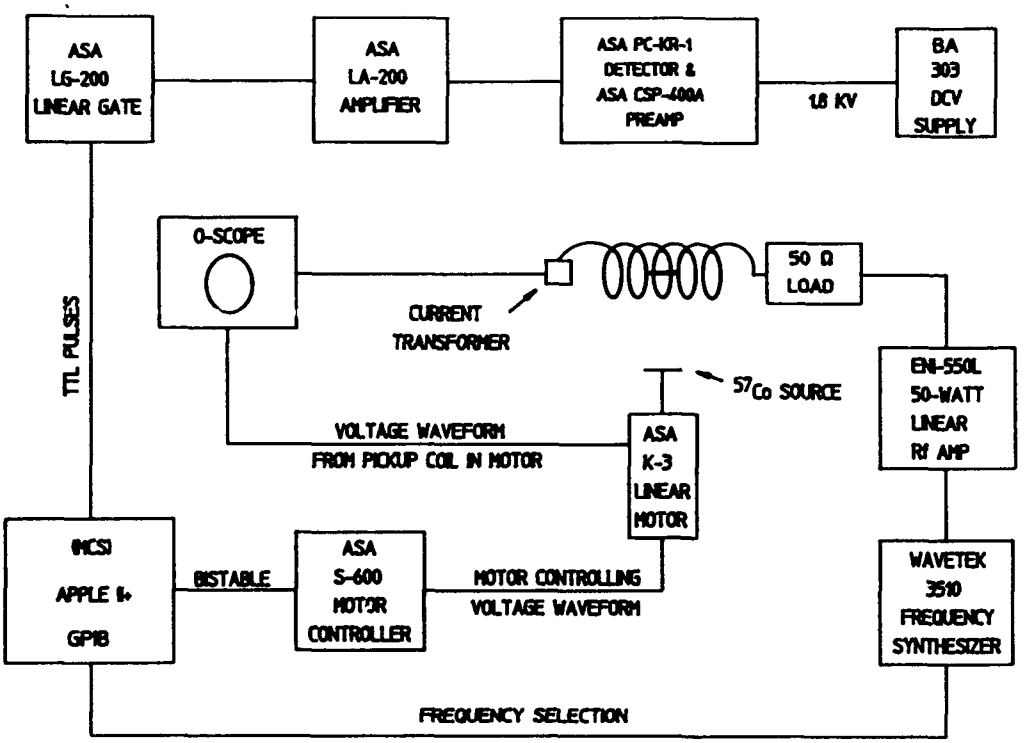

FIG. 1. Schematic representation of a frequency modulation spectrometer (FMS).

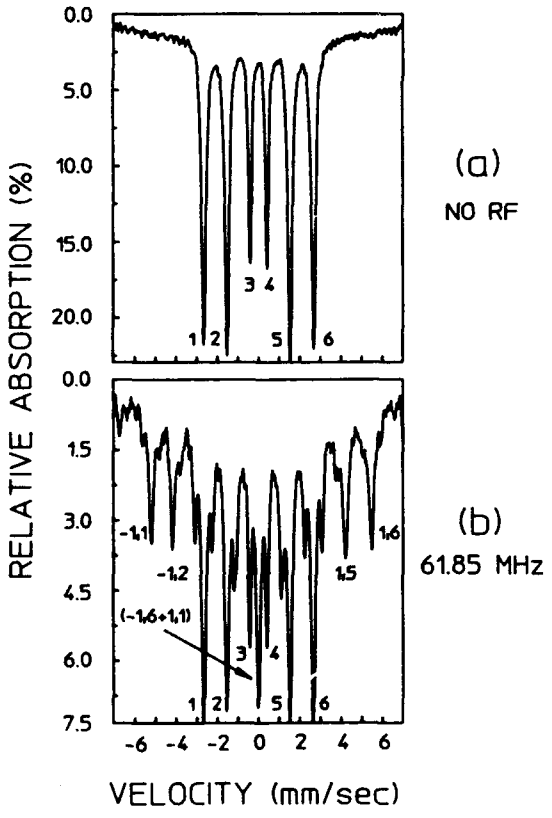

FIG. 2. (a) Mössbauer absorption spectrum of a 2.5-μm foil of iron isotopically enriched with 95.8% ^{57}Fe obtained with a ^{57}Co source in a Pd matrix. The six allowed magnetic dipole transitions are labeled 1 through 6 going from the lowest- to the highest-energy transition. (b) Spectrum of the same foil subjected to an oscillating radio-frequency field with a frequency of 61.85 MHz.

the incident gamma rays from the source will differ from the transition center of the absorber by the isomer shift, Δ. In a *FMS spectrum* the sidebands $+1;1$ would be observed at a frequency of $(61.85-\Delta)$ MHz while the sideband $-1;6$, would be detected at a frequency of $(61.85+\Delta)$ MHz. Therefore, the FMS spectrum will produce two absorption peaks as shown in Fig. 3(a), around 62 MHz with a separation equal to twice the isomer shift, 2Δ. If the Mössbauer source is given a constant velocity Doppler shift, the FMS spectrum will show two peaks near 62 MHz, separated by $2(\Delta+\delta)$, where δ is the Doppler shift of the incident gamma rays. In this way very precise measurements of the nuclear isomer shift can be obtained using FMS. The accuracy of the measurement is dependent upon the resolution and stability of the signal generator used to produce the rf magnetic field and the quality of the curve fitting routine used to fit the data.

I. THEORY

In a Mössbauer absorption spectrum taken in the velocity domain the isomer shift appears as a displacement of the centroid of the resonance pattern from the nominal zero position. The isomer shift is the result of an electric monopole interaction between the Mössbauer nuclei and its surrounding electrons. More specifically, the excited and ground-state nuclei differ in radius by a small yet significant amount ($\Delta R/R = -0.8\times10^{-3}$ in ^{57}Fe). It is this change in the radius of the nucleus which causes the electrostatic interaction between the nucleus and its surrounding electrons to change upon excitation of the nucleus. Therefore, if the Mössbauer nuclei in the source and absorber are in different

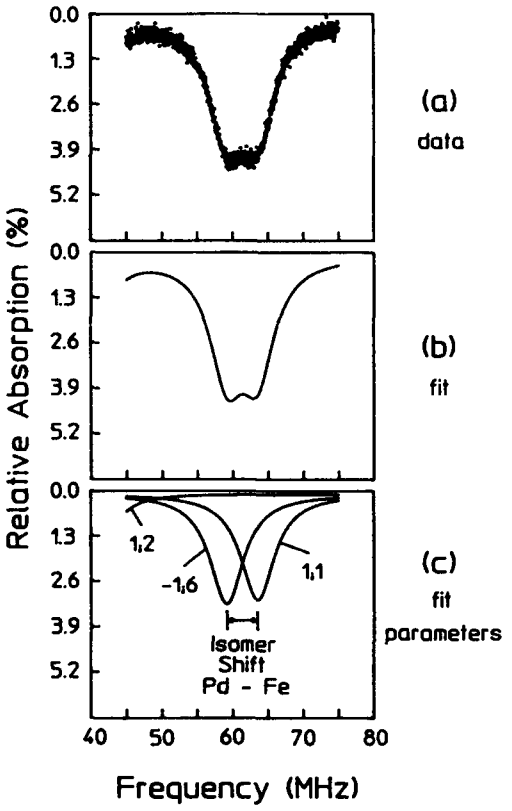

FIG. 3. (a) FMS spectrum of the same enriched foil. The two peaks represent sidebands + 1;1 and -1;6. The separation of these peaks is equal to twice the isomer shift of an iron foil relative to a ^{57}Co source in Pd. (b) Spectrum constructed from fitting the data of Fig. (a) using the Levenberg–Marquardt method for a nonlinear least-squares fit. (c) Spectral components resulting from the fitting procedure shown before summation.

chemical environments such that different electronic distributions are experienced, the isomer shift has been shown to be[16]

$$\Delta = (4\pi cZe^2R^2/5E_\gamma)[\rho_a(0) - \rho_s(0)](\Delta R/R) \text{ mm/s}, \quad (1)$$

where Z is the atomic number, e is the electronic charge, R is the effective nuclear radius, c is the velocity of light, $\rho_a(0)$ and $\rho_s(0)$ are the electronic charge densities at the nucleus for the source and absorber, and $\Delta R = R_{\text{excited}} - R_{\text{ground}}$.

In addition to the isomer shift displacement of the centroid of the resonance pattern there is also a temperature shift of the spectrum which arises due to time dilation resulting from the motions of the emitting and absorbing nuclei. This temperature shift is given by[17]

$$\delta E/E = C_p/2c^2 \quad (2)$$

where C_p is the specific heat of the material and c is the velocity of light. At room temperature, for iron the above expression yields $\delta E/E = -2.21\times10^{-15}$/°C. The experimentally observed value[18] is $-(2.09\pm0.24)\times10^{-15}$/°C. In the work being reported here the source and absorber were at essentially the same temperature. Therefore, the temperature shift was considered negligible.

The frequency, f_n, at which sidebands appear in a FMS spectrum is given by

$$f_n = (v - P_i + \Delta)(K/n), \quad (3)$$

where v (mm/s) is the Doppler velocity of the source, P_i (mm/s) is the position of the ith parent transition, Δ (mm/s) is the isomer shift, K(= 11.605 MHz/mm/s) is the conversion factor for the 14.4-keV gamma rays being detected and n is the order of the sideband of interest. Furthermore, it is known that the frequency displacement Ω of sidebands 1;1 and − 1;6 is given by

$$\Omega(1;1) = \Omega(-1;6), \quad (4)$$

with respect to the center of the resonance pattern. Therefore, for a stationary source and absorber we have

$$\Omega(1;6) - \Omega(1;1) = |\Omega(P_6) + \Delta| - |\Omega(P_1) + \Delta| = 2\Delta. \quad (5)$$

The absolute value of the Ω's are used since Eq. (3) can result in a negative frequency. However, the sign of f_n is merely a convention to indicate whether the sideband has a lesser or greater energy than its parent transition. The sign convention used for isomer shift measurements is that a negative isomer shift indicates the source has a higher nuclear energy level than the absorber.

II. DATA AND DISCUSSION

The isomer shift between a ^{57}Co source in a Pd matrix relative to an iron foil absorber was measured using FMS and found to have a value of $-(0.1869\pm0.003)$ mm/s compared to the literature value[19] of $-(0.185\pm0.02)$ mm/s. The ^{57}Co source in a Pd matrix had an initial activity of 25.4 mCi and was purchased from New England Nuclear. The absorber was a 1.5 cm×0.85 cm×2.5 μm foil of iron isotopically enriched with 95.8% ^{57}Fe. The rf signal was produced by a Wavetek signal generator, Model 3520, which was equipped with an IEEE-488 GPIB interface to allow the frequency to be automatically scanned. The resolution and stability of this signal generator are those previously stated. The rf signal from the Wavetek was amplified by an ENI-5506 50db linear amplifier and this amplified signal was then used to drive an inductor in which the absorber was mounted. A five-turn flat coil was constructed as the field induction coil with a separation of 4.5 mm between turns and a cross-sectional area of 20 mm^2. This led to a small inductance (< 1 μH) and a minimum capacitance between turns while producing a reasonable field intensity (~4G) at 50 W. This inductor was then mounted in series with a 48-Ω noninductive load to match the impedance of the amplifier output and cable. The result was a circuit having a reasonably broad band tunability and a Q of approximately one. The heat produced in the absorber by eddy currents was minimal for the rf powers employed. However, in critical measurements of the isomer shift using FMS, this minimal rf heating of the absorber can be eliminated by simultaneously pulsing the rf and directing a cool flow of nitrogen over the absorber.

The data from which the fitted parameters were obtained is shown in Fig. 3(a). The data was fit to seven parameters using the Levenberg–Marquardt method[20] for a nonlinear least squares fit. In the fitting procedure, it was assumed that the spectra were composed of three sidebands; the − 1;6, + 1;1, and − 1;5. It was further assumed that

the sidebands had a uniform linewidth. Figure 3(b) shows the spectrum constructed from the fit parameters and Fig. 3(c) shows the spectral components before summation. The standard deviation of the fit parameters was obtained from the covariance matrix derived from the fitting procedure and are valid only with the assumption of normally distributed measurement error.

As can be seen from this example, FMS is a high-resolution Mössbauer technique which provides a direct measurement of rf sideband positions and intensities without interference from the parent transitions. Furthermore, from an FMS spectrum, information can be obtained without mechanical tuning about the linewidth or positions of Zeeman split energy levels in addition to accurate measurement of the nuclear isomer shift. Recently[14] it has been demonstrated how to excite large-scale magnetic phase modulation effects in paramagnetic materials by exciting magnetostatic spin waves in the sample under study. With this refinement frequency modulation spectroscopy can now be extended to the study of paramagnetic materials. Then, FMS can provide a viable alternative for precision isomer shift measurements in a variety of environments.

ACKNOWLEDGMENTS

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Thermomechanical Frequency Modulation of γ Radiation

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When a radio-frequency current is passed through a paramagnetic Mössbauer absorber containing ^{57}Fe, sidebands are generated on the 14.4-keV transition at $\omega_\gamma \pm 2n\omega$, where $n = 1,2,\ldots$, and ω is the applied frequency. The new aspect is that only sidebands of even orders appear. These sidebands are shown to be the result of thermomechanical oscillations induced by the rf current.

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Frequency, phase, and amplitude modulation of γ rays has been of interest ever since the discovery of the Mössbauer effect made such studies possible.[1-5] When considering these phenomena, there is an immediate distinction to be made between Doppler-type frequency shifts caused by microscopic motion of the nuclei inside sources or absorbers[2,3] and modulation resulting from a direct interaction of periodically varying magnetic fields with the nuclear moments. The latter can result either in a "dressing" of nuclear states[5] or in rf collapse of hyperfine spectra.[6,7] The modulation of nuclear states[8] employed either internal magnetic fields in the case of ^{57}Fe or external alternating fields applied directly to longer-lived nuclear states. In contrast, frequency and phase modulation originating from motion was generally achieved either by rocking source or absorber with a piezoelectric transducer[2] or by magnetostrictive motion,[3] because of its convenience for ^{57}Fe.

To our knowledge, a conceptually simple form of motion, namely, thermal expansion, was never exploited for modulating γ rays. In this Letter we want to point out that periodic thermomechanical motion leads to sidebands of *only even orders*, distinctly different from those generated by alternating hyperfine fields or by magnetostrictive oscillations. To demonstrate this difference we will contrast the influence of an alternating heating current through the absorber foil on the Mössbauer spectrum against the effect of modulating the phases of nuclear states by alternating hyperfine fields due to spin waves.[9]

The arrangements in which sidebands were excited on the 14.4-keV transition of ^{57}Fe in this work are shown in Fig. 1. In one variant the rf current is passed directly through a stainless-steel (SS) foil in which the absorption takes place [Fig. 1(b)] and in the other current flows through an Al strip in contact with a ferromagnetic Ni pad which separates the Al strip from the stainless absorber [Fig. 1(a)]. In the latter scheme the time-varying magnetic field in the Al foil generates spin waves in the Ni that are transferred to the stainless absorber.[9] This arrangement is convenient since no coils or resonantly tuned circuits are needed to launch the spin waves and the magnetic field generated by the current in the Al strip can be easily calculated. The resulting spectra are seen in Fig. 2. The spectrum excited with the Ni pad

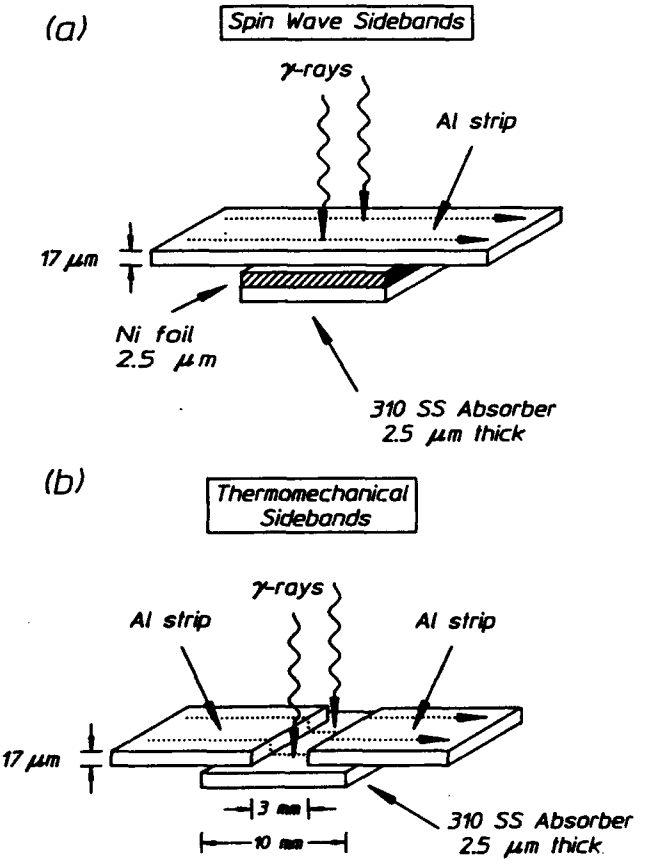

FIG. 1. Schematic representation of the arrangement used to excite (b) thermomechanical sidebands in comparison to (a) the typical scheme for producing spin-wave sidebands. The path of radio-frequency current is shown by the dotted arrow.

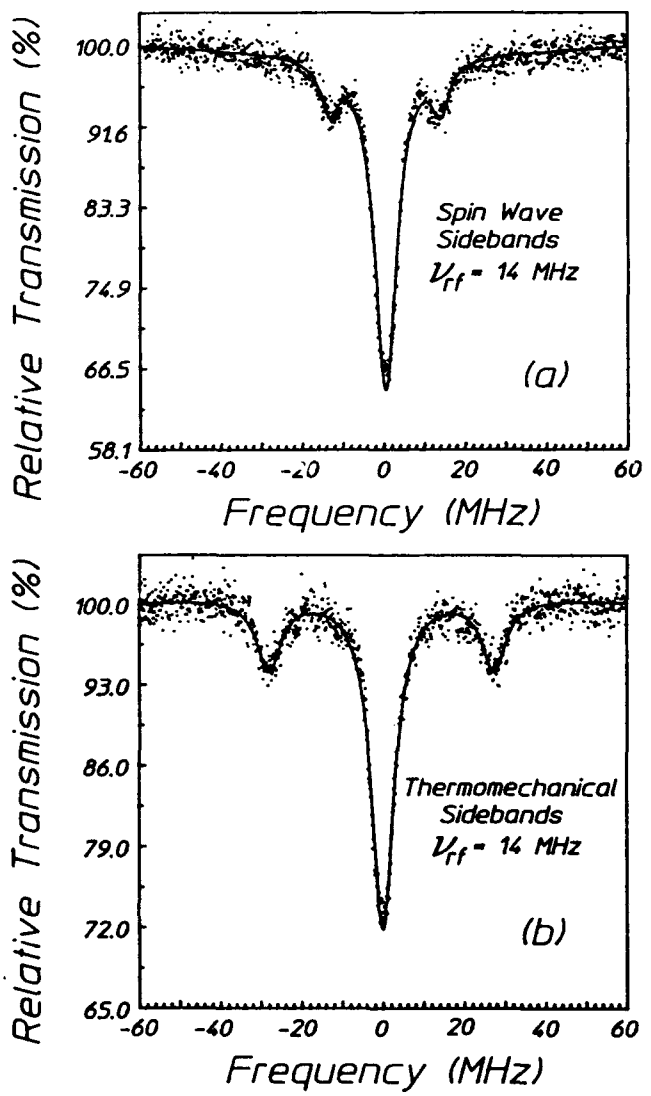

FIG. 2. Absorption data obtained with the comparative arrangement of Fig. 1. The spectra were obtained by using constant acceleration between source and absorber to produce a Doppler shift of energies by the amounts shown on the abscissa in frequency units. The positions of the spectra correspond to the arrangements shown in Fig. 1 used to excite them. In *both cases the driving frequency was 14 MHz.*

shows normal sidebands of first order [Fig. 2(a)], while the one obtained with the arrangement of Fig. 1(b) has only sidebands of second order [Fig. 2(b)]. With the mounting of Fig. 1(b), higher even orders are excited (Fig. 3) if the current is increased or the frequency decreased.

Several tests indicated that sidebands excited with the intermediate Ni foil are simply spin-wave sidebands resulting from the periodic magnetization of the Ni by the currents flowing through the adjacent Al strip. The magnetic nature of these sidebands was confirmed by experiments in which the Al foil was separated from the Ni drivers by an insulating layer. Sidebands were still produced in the stainless foil with insulating layers of about

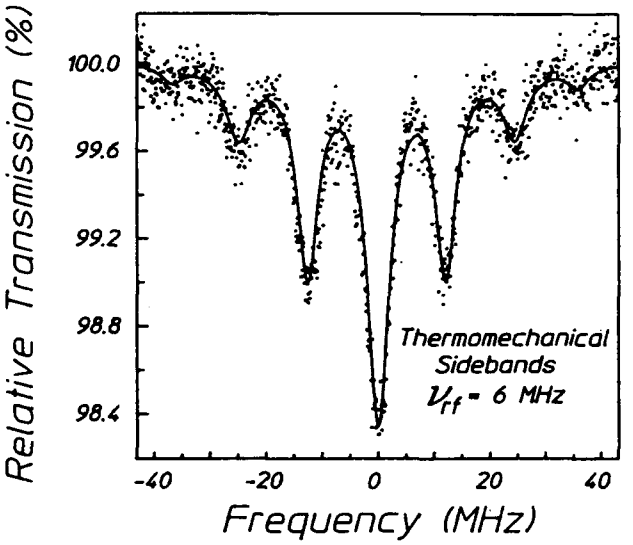

FIG. 3. Absorption data obtained with the arrangement of Fig. 1(b) for a driving frequency of 6 MHz. The spectra were obtained by using constant acceleration between source and absorber to produce a Doppler shift of energies by the amounts shown on the abscissa in frequency units.

100 μm thickness. The phenomenology in this case was the same as that for the spin-wave excitation reported earlier.[9] In particular, those effects were not sensitive to mounting or to any mechanical damping of spurious vibrations which might have been present in the foil.

For cases in which there is no static magnetic field[5] or in which the modulation is parallel to the static field,[8] the effect of a time-varying component of magnetic field $H_0 f(t)$ upon an eigenstate of a nucleus, $\Psi^{(0)}_{a,m}$, can be written as

$$\Psi_{a,m} = e^{-i\phi_a(t)}\Psi^{(0)}_{a,m}, \tag{1}$$

where $\phi_a(t)$ is the modulation angle of the phase,

$$\phi_a(t) = m\omega_a \int_0^t f(t')dt'. \tag{2}$$

The Larmor frequency ω_a is given by $\mu_N g_a H_0/\hbar$, where μ_N is the nuclear magneton, and g_a the gyromagnetic ratio for the ath excited or ground state of the nucleus, and m is the magnetic quantum number of the eigenstates. As can be seen, the effect of the field is only to introduce an additional time-dependent factor modifying the time dependence already present in the wave function $\Psi^{(0)}_{a,m}$. However, it is the difference in phase modulation between the ground state g and an excited state e that is observed during an absorption transition, and the Fourier components of $\phi_e(t) - \phi_g(t)$ are manifested as sidebands. In this case all orders are expected.[1,3,5,8,9]

Helisto *et al.*[10] have given another nuclear phase associated with the position of the nucleus in a medium which can be written as

$$\phi_x(t) = 2\pi x(t)/\lambda_\gamma, \tag{3}$$

where λ_γ is the wavelength of the Mössbauer radiation. This phase reflects the spatial origin of the γ-ray emission, and therefore does not affect the internal phase of the nuclear state shown in Eq. (2). It is the phase which is produced by vibration or other mechanical effects like thermomechanical motion.

To describe thermomechanical perturbations, we assume that the heating associated with the current flowing through a localized portion of the foil causes a linear expansion

$$\Delta x = \alpha\lambda_1(\Delta T)/2\,, \tag{4}$$

where α is the thermal-expansion coefficient and λ_1 is the wavelength for a longitudinal-acoustic vibration with velocity v_1. Since any expansion of the foil can propagate only at the velocity of sound, contributions to the displacement of a particular nucleus will be out of phase if they start from a part of the foil more remote than $\lambda_1/2$, so the tacit assumption has been made that only the region within $\lambda_1/2$ around a nucleus can contribute to the phase change by thermal expansion. Effects from regions farther away would tend to excite an acoustic wave that would not be coherent with the one building within the first $\lambda_1/2$ adjacent to the nucleus.[11]

In the arrangement of Fig. 1, the volume V having resistance R of the foil was heated by a current passing along a length l through a cross-sectional area A,

$$i = i_0\cos(\omega t)\,, \tag{5}$$

and was cooled by conduction through the thin dimension into a microscope cover slide. Joule heating due to the rf current forces the surface of the foil to undergo a periodic temperature variation given by

$$\Delta T = \frac{i_0^2 R}{2V\rho C}t + \frac{i_0^2 R}{4V\rho C\omega}\sin(2\omega t)\,, \tag{6}$$

where $V\rho C$ is the heat capacity of the volume.

When heat dissipation is taken into account, the first term of Eq. (6) is modified by the specifics of the geometry to give an asymptotic temperature rise to an elevated average in the limit of longer times. This asymptotic phase change associated with the thermal expansion of the foil to the steady-state temperature is of no importance in this context and will be dropped. Of interest is the second term which is unaffected by linear processes of dissipation and yields the periodic temperature variation of the foil. Substituting this second term of Eq. (6) into Eq. (4) and then into Eq. (3) gives

$$\phi_x(t) = (2\pi x_0/\lambda_\gamma)\sin(2\omega t)\,, \tag{7}$$

where

$$x_0 = \alpha i_0^2 R\lambda_1/8V\rho C\omega\,. \tag{8a}$$

Substituting $i_0 = j_0 A$, $V = Al$, and $\lambda_1 = 2\pi v_1/\omega$, $R = rl/A$, where j_0 is the current density and r is the resistivity of the foil gives the form

$$x_0 = (\pi\alpha r v_1/4\rho C)(j_0/\omega)^2\,, \tag{8b}$$

in which the dominant physical variables are readily apparent.

For the enriched 310 stainless-steel foils used in these experiments a current density of $j_0 = 1.4\times10^9\ \mathrm{A\,m^{-2}}$ at a frequency of $\omega/2\pi = 10$ MHz would give a magnitude of the phase change in Eq. (7) of $\phi_0 = 0.5$. The corresponding amplitude of the oscillation is approximately 0.1 Å and the temperature fluctuations according to Eq. (6) would be only 1.6×10^{-3} K. Such a small value is only about 3×10^{-6} of the Debye temperature and so could not contribute any additional amplitude modulation to the recoilless fraction of events.

The quantity ϕ_0 is equivalent to the modulation index of a carrier wave and is a measure of the amount of spectral intensity appearing in the lowest-order sideband. This can be appreciated by considering that the nucleus emits a γ wave in the x direction described by

$$\bar{E} = \bar{E}_0\exp\{i[kx - \omega_\gamma t + \phi_0\sin(2\omega t)]\}\,. \tag{9}$$

For this problem a useful representation of the Jacobi-Anger expansion is

$$\exp[iz\sin(2\omega t)] = \sum_{n=-\infty}^{\infty} J_n(z)\exp(i2n\omega t)\,. \tag{10}$$

Substituting this into Eq. (9) gives for the propagating γ wave

$$\bar{E} = \sum_{n=-\infty}^{\infty} \bar{E}_0 J_n(\phi_0)\exp\{i(kx - [\omega_\gamma - 2n\omega]t)\}\,. \tag{11}$$

The interpretation of Eq. (11) is that the γ ray is a sum of components with frequencies $\omega_\gamma \pm 2n\omega$ and relative amplitudes of $J_n(\phi_0)$. Notice that the excitation of the even-order sidebands is a consequence of Joule heating. Since intensity is basically E^2, the relative intensity I_m of the mth sideband is $I_m = I_0 J_n^2(\phi_0)$, again where ϕ_0 is the modulation index.

While the required value of current density is large, it is consistent with the difficulty in performing this experiment. The problems of insuring a uniform j_0 in the foil limited quantitative comparison to the predictions, but sidebands such as shown in Fig. 2(b) required on the order of 10 A flowing through Al strips 2 mm wide contacted to the 2.5-μm-thick stainless-steel absorber. At 14 MHz, such as used to obtain those particular data in Fig. 2(b), the expected modulation index is about 0.5. The important feature is that Eq. (11) predicts only even-order sidebands and that is what is seen in Figs. 2(b) and 3. The data shown there represent the first time thermomechanical sidebands have been excited in Mössbauer spectroscopy and, moreover, the first time sidebands have been excited by any means without odd orders appearing in addition.

It was also found that the observations of ther-

momechanical sidebands were laterally restricted to the region of the current path. They were not readily transported away from the heat source in the stainless absorber, a characteristic in complete contrast to the ease with which spin waves could propagate in the same medium. This is entirely consistent with the model since the source term for the Joule heating is effective only in the source area and cannot propagate more than the thermal diffusion length, $\delta_T = (2D/\omega)^{1/2}$, where D is the thermal diffusivity of the material. For our stainless-steel absorber δ_T is about 0.3 μm, almost an order of magnitude smaller than the thickness. With regard to the depth penetration the same consideration applies and we expect the sideband intensity to drop correspondingly with increasing absorber thickness. The thermomechanical origin of the even-order sidebands was further supported by the general observations that it was very easy to damp these sidebands mechanically.

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Quantum Interference in the Mössbauer Spectrum of ^{57}Fe Induced by Radiofrequency Magnetic Fields

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Abstract

In this paper we show that the Mössbauer spectrum of ^{57}Fe under the influence of an applied radiofrequency magnetic fields should display a characteristic structure due to quantum interference, in addition to the conventional resonances. It is found that at certain gamma-ray energies the amplitudes for the various possible interfering channels cancel each other, so that the transition probability actually vanishes at those energies.

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If a physical process proceeds from an initial to a final state through a number of intermediate states, the quantum mechanical amplitude of that process will be in general the sum of the amplitudes of each of the available channels. When this resultant amplitude is squared to obtain the observable probabilities the interferences appear that are the signatures of the quantum nature of the process.

While the two-slit scattering experiment, either with electrons or with photons, can be regarded as the paradigm of quantum mechanics, the purpose of this paper is to bring into evidence a remarkable example of quantum interference in the case of multiphoton transitions involving gamma-ray quanta and radiofrequency photons. Thus, it will be shown that the Mössbauer spectrum of ^{57}Fe nuclei under the influence of an applied radiofrequency magnetic field displays a characteristic structure in addition to the conventional resonances. In particular, it is found that at certain gamma-ray energies the amplitudes for the various possible channels cancel each other so that the transition probability actually vanishes at those energies.

In order to determine the shape of the Mössbauer gamma-ray spectrum we shall use the published formulas[1] for the intensity of the nuclear multiphoton transitions for ^{57}Fe nuclei driven by static and radiofrequency magnetic fields. Any static field produces a Zeeman splitting of the hyperfine levels, so that six components appear for the transition between the lowest excited state of 14.4 keV and the ground state. The addition of radiofrequency fields produces sidebands to the parent transitions, as previously reported in literature.[1-3]

For convenience, we shall denote the lines of the unperturbed gamma-ray spectrum by a, b, c, d, e, f, in the order of decreasing energy and the intensity of the transitions by I. Furthermore, we shall designate the sidebands to the right and to the left of the parent transition with the subscripts + and - respectively, and distinguish the cases of linear,

circularly-right and circularly-left polarizations of the applied radiofrequency field with the aid of the superscripts L, CR, and CL, respectively.

Analyzing the case of linear polarization of the applied radiofrequency magnetic field first, we shall consider that a static magnetic field B_o^h producing a splitting of 45.43 MHz in the ground state of ^{57}Fe nucleus is applied along the z-axis, while the oscillating component of the magnetic field is oriented along the x-axis. If the gamma-ray beam is perpendicular to the xz plane, and if we assume that the incident gamma-ray beam is unpolarized, it can be shown[1] that the ratio of the intensity of the first-order sidebands of the line a to the intensity of the parent line is

$$\frac{I_{a\pm}^{(L)}}{I_a} = \frac{1}{4}\left(\frac{B_{rf,x}}{B_o^h}\right)^2 \frac{\omega_e^2}{(\omega_{rf}\pm\omega_e)^2} \quad , \tag{1}$$

where $B_{rf,x}$ is the amplitude of the radiofrequency field, B_0^h the static hyperfine field, ω_{rf} is the frequency of the oscillating field and ω_e = 25.96 MHz. The conventional profiles described in Eq. 1 are shown in Fig. 1.

The relative intensities of the sidebands to the parent transition b can be shown to be

$$\frac{I_{b\pm}^{(L)}}{I_b} = \left(\frac{B_{rf,x}}{B_o^h}\right)^2 \left[-\frac{3}{8}\frac{\omega_e}{-\omega_e\pm\omega_{rf}} + \frac{1}{4}\frac{\omega_e}{\omega_e\pm\omega_{rf}} - \frac{1}{8}\frac{\omega_g}{\omega_g\pm\omega_{rf}}\right]^2 \quad . \tag{2}$$

Unlike the monotonic wings of the conventional profiles, it is apparent from Fig. 2 that the transition intensity $I_{b-}^{(L)}$ has a minimum at ω_{rf} = 4.4 MHz.

The most interesting pattern occurs, however, for the sidebands of the parent line c. The relative intensities of the sidebands are for this case given by

$$\frac{I_{c\pm}^{(L)}}{I_c} = \left(\frac{B_{rf,x}}{B_o^h}\right)^2 \left[\frac{\omega_e}{-\omega_e \pm \omega_{rf}} + \frac{1}{2}\frac{\omega_g}{\omega_g \pm \omega_{rf}}\right]^2 \quad . \qquad (3)$$

From Eq. 3 it is apparent that due to a cancellation of amplitudes, the transition intensity $I_{c-}^{(L)}$ vanishes at the frequency $\omega_{rf} = \omega_e\omega_g/(\omega_g + 2\omega_e)$, or $\omega_{rf} = 12.1$ MHz, as shown in Fig. 3.

The observation of these structures in the sideband intensity may prove difficult in the case of a linear polarization of the applied radiofrequency field because of the overlapping of sidebands to the various parent lines. Less complex spectra, however, are expected to be obtained in the case of circularly-polarized radiofrequency fields. We now consider that the static magnetic field B_0^h is oriented along the z-axis but assume a circularly-left polarized radiofrequency field rotating in the xy-plane. The direction of the gamma-ray beam is perpendicular to the xy plane. In this case, $I_{a\pm}^{(CL)} = 0$, $I_{b\pm}^{(CL)} = 0$, while

$$\frac{I_{b+}^{(CL)}}{I_a} = \frac{1}{3}\left(\frac{B_{rf}}{B_o^h}\right)^2 \frac{\omega_e^2}{(\omega_{rf}+\omega_e)^2} \quad , \qquad (4a)$$

$$\frac{I_{b-}^{(CL)}}{I_a} = \frac{1}{12}\left(\frac{B_{rf}}{B_o^h}\right)^2 \left[\frac{3\omega_e}{\omega_{rf}+\omega_e} - \frac{\omega_g}{\omega_g-\omega_{rf}}\right]^2 \quad , \qquad (4b)$$

where B_{rf} is the magnitude of the rotating magnetic field. We have compared the intensities $I_{b\pm}^{(CL)}$ to the parent intensity I_a since in this case $I_b = 0$. The pattern of the lines $I_{b\pm}^{(CL)}$ is shown in Fig. 4. In the case of $I_{b-}^{(CL)}$, the transition intensity vanishes at $\omega_{rf} = 2\omega_e\omega_g/(3\omega_e + \omega_g)$, or $\omega_{rf} = 19.1$ MHz, due to a cancellation of the amplitudes. Similar effects can be observed in the pattern of the transition intensities for circularly-right polarized magnetic fields.

The results of the present work suggest the importance of an experimental investigation of the multiphoton Mössbauer spectrum of ^{57}Fe at frequencies

off-resonance. Moreover, while the two-slit scattering experiment with electrons or photons constitute the paradigm of quantum mechanics, the observation of the spectral structure described in this paper would provide equally compelling evidence for the process of interference of quantum mechanical amplitudes, and in this case at the nuclear level.

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FIGURE CAPTIONS

Figure 1. Intensity of the sidebands $I_{a+}^{(L)}/I_a$ (dotted line) and $I_{a-}^{(L)}/I_a$ (solid line), in units of $(B_{rf,x}/B_o^h)^2$, for a linearly-polarized radiofrequency magnetic field as a function of the frequency ω_{rf} of this field.

Figure 2. Intensity of the sidebands $I_{b+}^{(L)}/I_b$ (dotted line) and $I_{b-}^{(L)}/I_b$ (solid line), in units of $(B_{rf,x}/B_o^h)^2$, for a linearly-polarized radiofrequency magnetic field as a function of the frequency ω_{rf} of this field. Due to quantum interference, $I_{b-}^{(L)}$ displays a characteristic structure around ω_{rf} = 4.4 MHz.

Figure 3. Intensity of the sidebands $I_{c+}^{(L)}/I_c$ (dotted line) and $I_{c-}^{(L)}/I_c$ (solid line), in units of $(B_{rf,x}/B_o^h)^2$, for a linearly-polarized radiofrequency magnetic field as a function of the frequency ω_{rf} of this field. Due to quantum interference, $I_{c-}^{(L)}$ vanishes at ω_{rf} = 12.1 MHz.

Figure 4. Intensity of the sidebands $I_{b+}^{(CL)}/I_a$ (dotted line) and $I_{b-}^{(CL)}/I_a$ (solid line), in units of $(B_{rf}/B_o^h)^2$, for a circularly left polarized radiofrequency magnetic field as a function of the frequency ω_{rf} of this field. Due to quantum interference, $I_{b-}^{(CL)}$ vanishes at ω_{rf} = 19.1 MHz.

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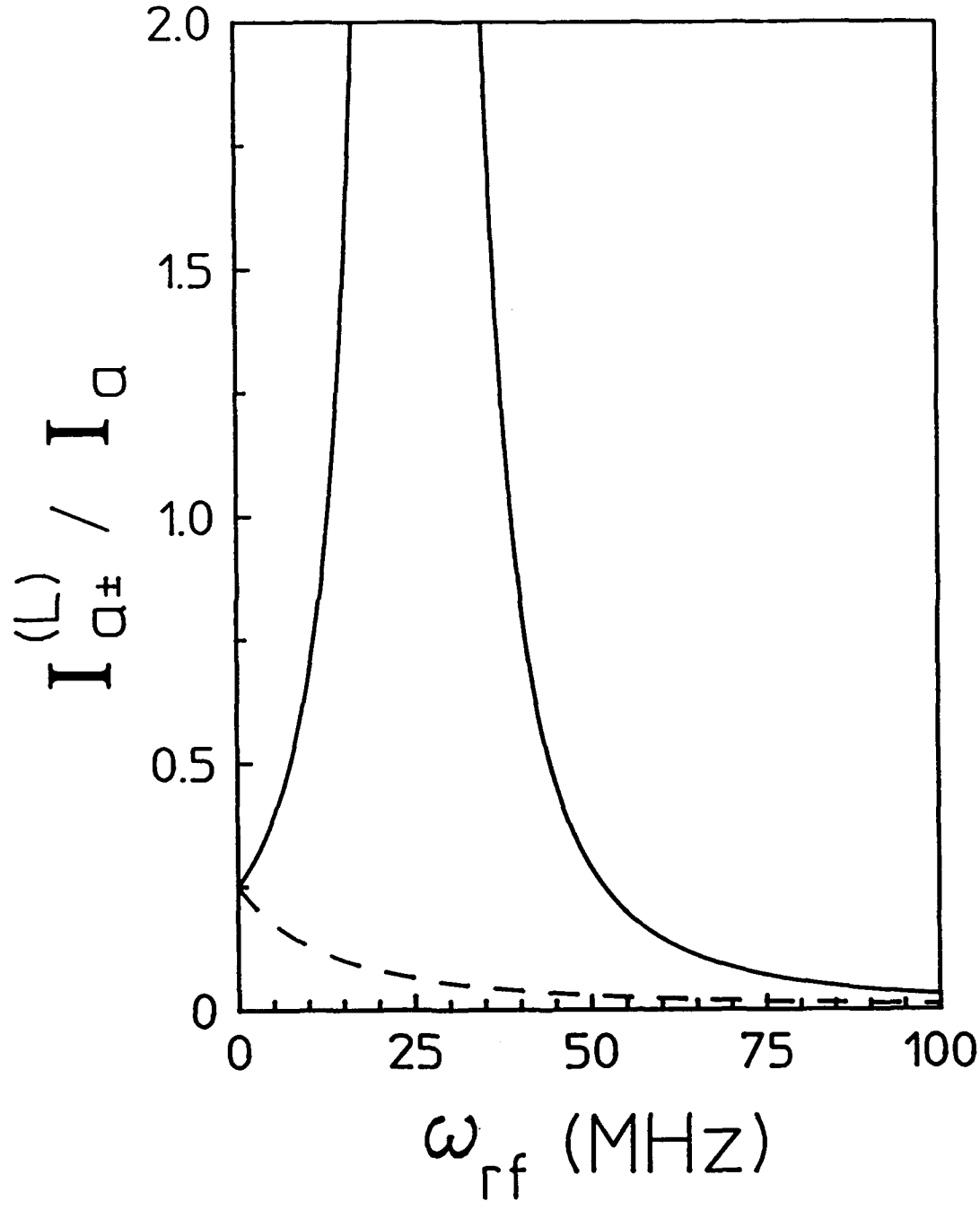

Fig 1

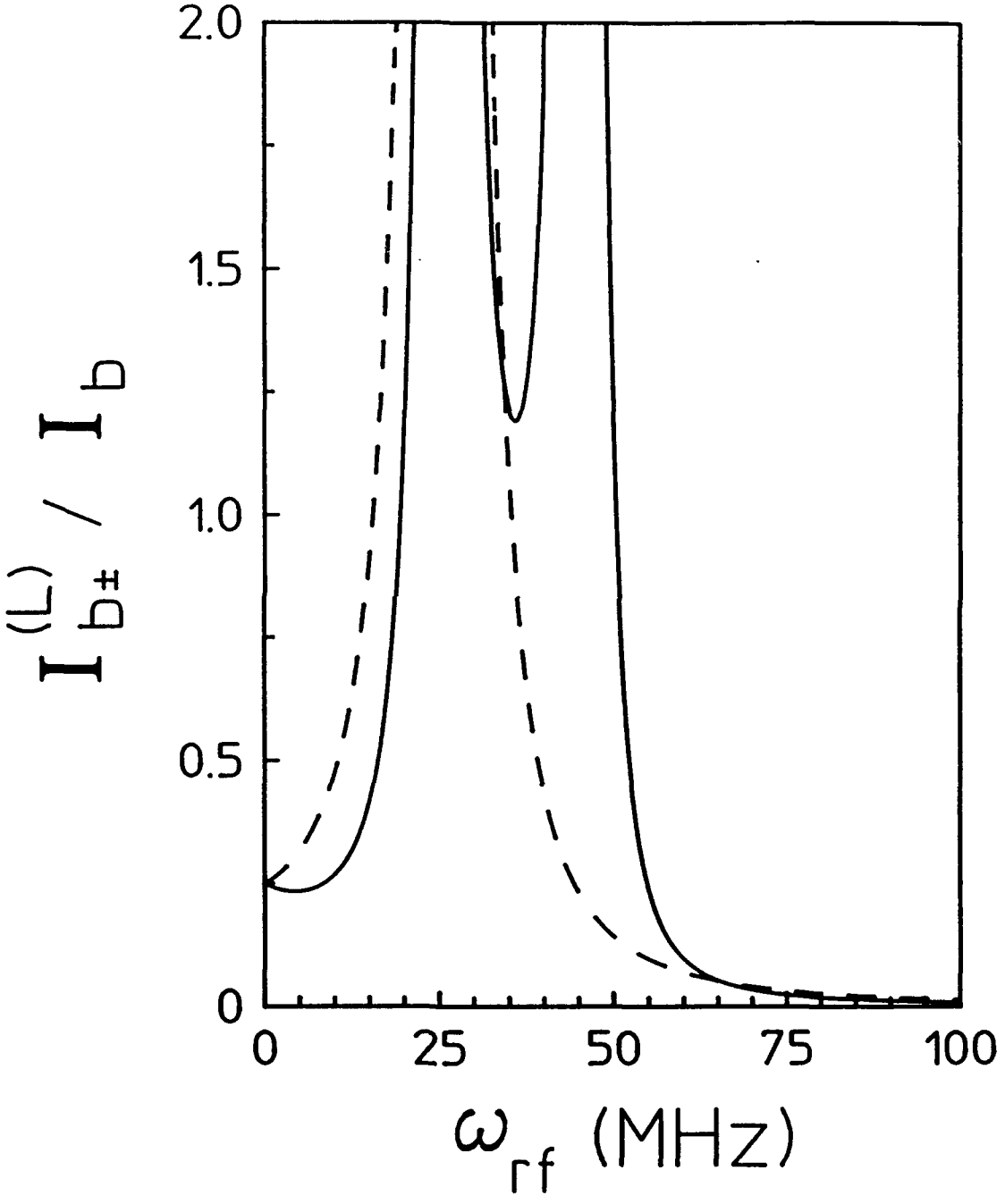

Fig 2

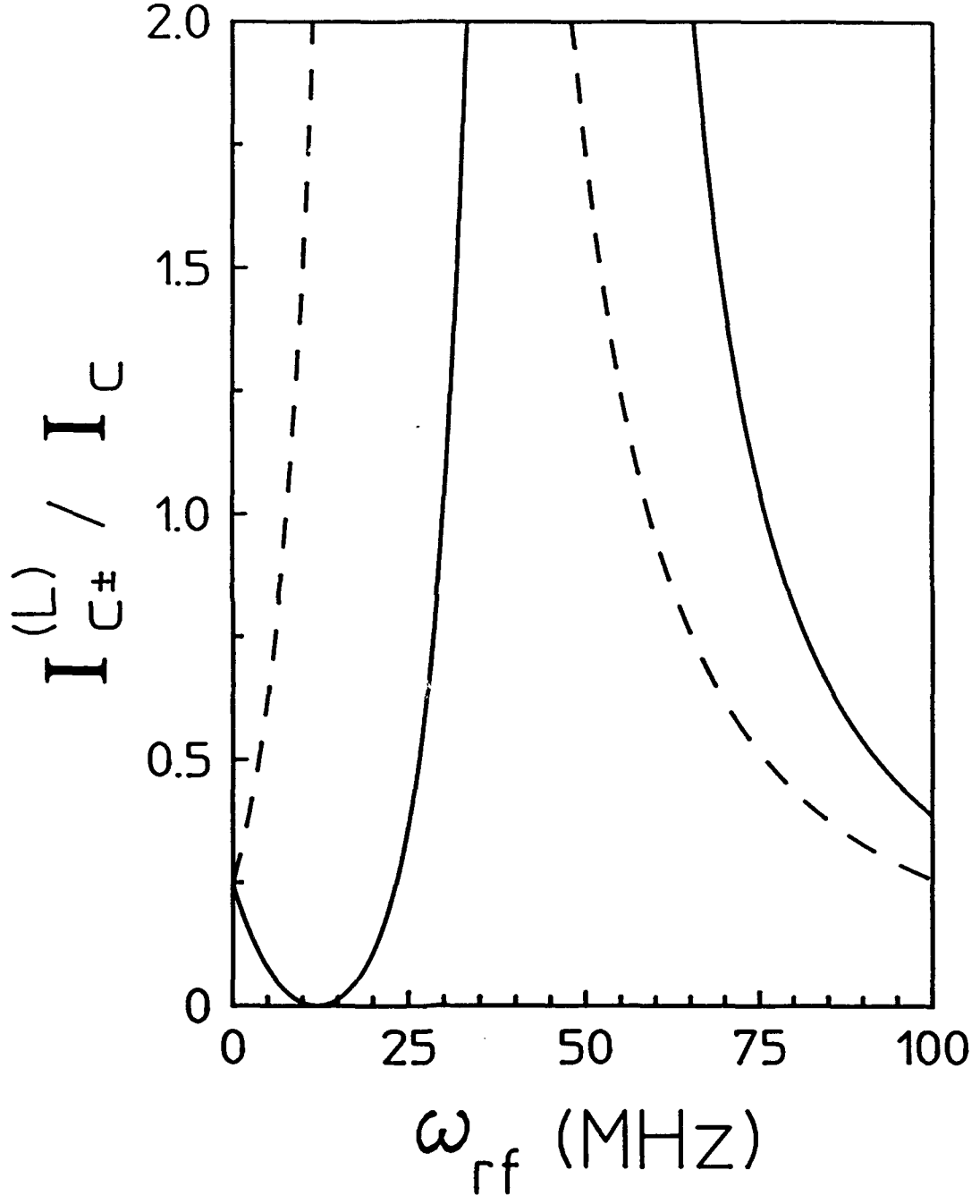

Fig 3

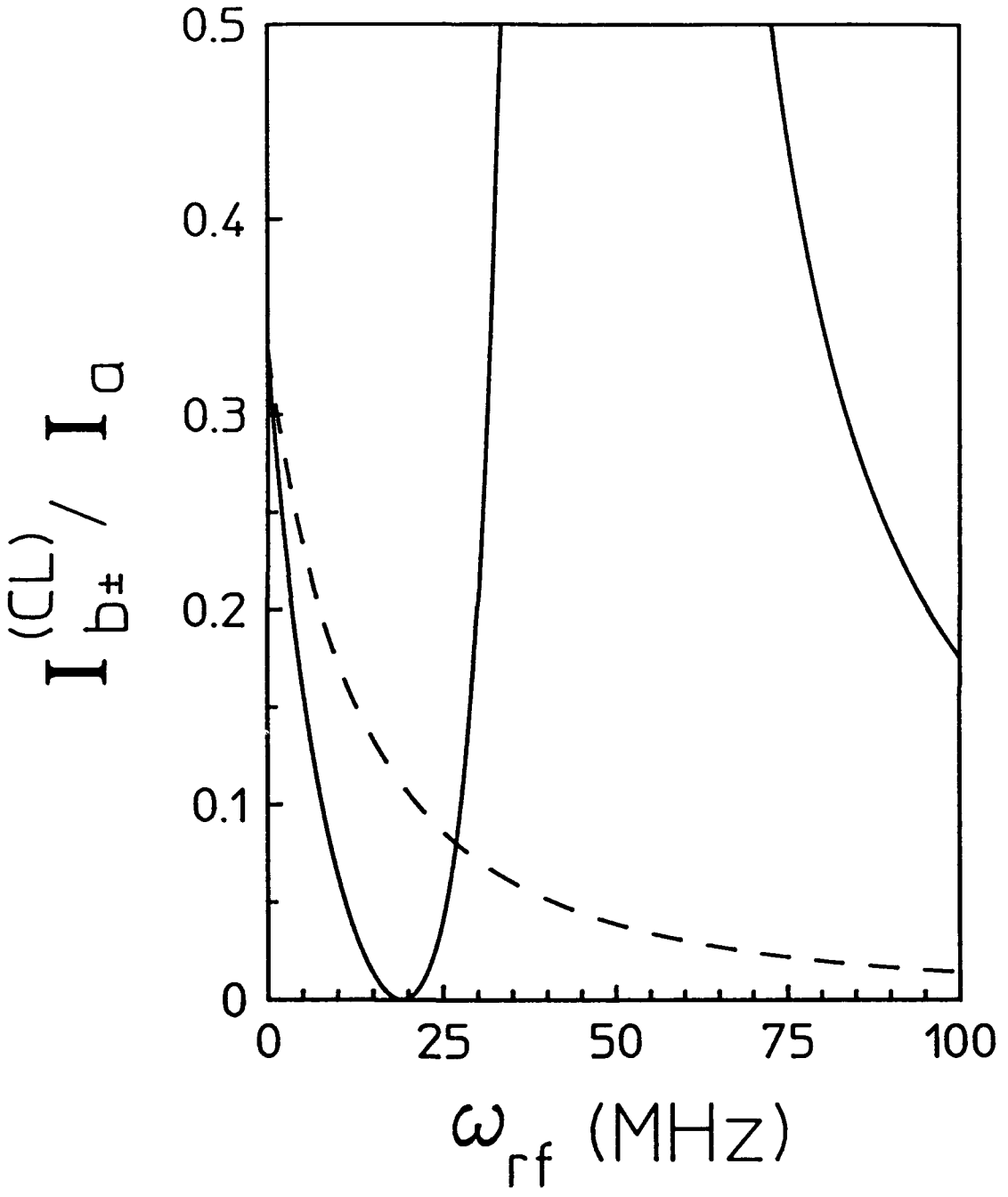

Fig 4